Dynamically Induced Excitonic Instability in Pumped Dirac Materials

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Driven and non-equilibrium quantum states of matter have attracted growing interest in both theoretical and experimental studies in condensed matter physics. Recent progress in realizing transient collective states in driven or pumped Dirac materials (DMs) is reviewed herein. In particular, the focus is on optically pumped DMs which are a promising platform for transient excitonic instabilities. Optical pumping combined with the linear (Dirac) dispersion of the electronic spectrum offers a knob for tuning the effective interaction between the photoexcited electrons and holes, and thus provides a way of reducing the critical coupling for excitonic instability. As a result, a transient excitonic condensate could be achieved in a pumped DM while it is not feasible in equilibrium. A unifying theoretical framework is provided for describing transient collective states in 2D and 3D DMs. The experimental signatures are described and numerical estimates of the size of the dynamically induced excitonic gaps and the values of the critical temperatures for several specific systems, are summarized. In addition, general guidelines for identifying promising material candidates are discussed. Finally, comments are provided regarding recent experimental efforts in realizing transient excitonic condensate in pumped DMs, and outstanding issues and possible future directions are outlined.

1. Introduction

The last decade has seen an increased interest in generating and controlling dynamical and non-equilibrium states of matter. Such states can be induced by interaction with a time-dependent drive such as electromagnetic fields or coherent lattice vibrations. Prominent recent examples include novel dynamical states in periodically driven systems, such as Floquet topological insulators[1–6] and time crystals,[7–10] nonlinear optical and transport effects in graphene and related materials,[11] phonon-driven Floquet states,[12] light-induced superconductivity,[13,14] and transient, or non-equilibrium, exciton condensate in pumped semimetals[15,16] and semiconductors.[17–19] Another example is intrinsically dynamical orders such as odd-frequency, or Berezinskii, superconducting pairing,[20,21] which involves correlations non-local in time and naturally manifests in the time domain, see also related paper in this volume.[22]

Progress in investigating the dynamical states is fueled by the development of time-sensitive pump-probe techniques such as time- and angle-resolved photoemission spectroscopy (trARPES) and time-resolved terahertz (THz) spectroscopy, which are able to probe the non-equilibrium dynamics of electronic states at ultrafast (subpicosecond) timescales. Ultrafast spectroscopy is currently used to study quantum materials, that is, materials where strong quantum correlations lead to unusual emergent properties such as unconventional orders and topology.[23,24] Such studies may provide insights into the physics of quantum materials and reveal novel dynamically-induced states that are inaccessible in equilibrium.

In this contribution, we discuss recent efforts toward the realization of transient collective states in a particular class of quantum materials, known as Dirac materials (DMs).[25] DMs are characterized by nodes and a linear, Dirac-like, dispersion of the quasiparticle spectrum. This is a growing class of materials encompassing high-temperature $d$-wave superconductors,[26] superfluid $^3$He,[27] graphene,[28] topological insulators (TI),[29,30] Dirac (DSM)[31] and Weyl semimetals (WSM),[32,33] Dirac nodal line semimetals[34,35] and bosonic DMs.[36,37]

Due to the great potential of DMs for high-performance optoelectronic devices,[38,39] non-equilibrium dynamics of DMs has emerged as an important research topic. Many studies have focused on the interplay between light and Dirac states[40–42] and, in particular, on optical generation and control of spin-polarized currents on TI surfaces.[43] Ultrafast spectroscopies have been used extensively to study electron dynamic in 2D DMs, for example, graphene[44–52] and 3D TIs.[53,54] Ultrafast dynamics of the recently discovered 3D DMs, that is, DSM and WSM, is a less explored topic but pump-probe studies similar to 2D DMs have
recently appeared in the literature.[55–61] In particular, relaxation dynamics was studied in TaAs,[57,59] NbP[59] and NbAs[60] WSMs, and in Cd₃As₂ DSM.[61,62]

An important result of the pump-probe studies in DMs is the possibility of creating a broadband population inversion.[46,63] In the context of optical pumping, population inversion is a situation when after initial rapid thermalization two independent Fermi-Dirac distributions are established, with distinct chemical potentials for the non-equilibrium electron and hole populations (see Figure 1). Population inversion is crucial for realization of broadband THz lasing in DMs.[38] According to the most optimistic experimental reports, population inversion in graphene can be sustained on the timescale of 100 – 200 fs.[46,68-71]

More recently, long-lived optically excited states have been demonstrated in 3D TIs with lifetimes ranging from few picoseconds to hundreds of picoseconds, for example, $\tau \approx 3$ ps in Sb$_2$Te$_3$[53] and $\tau \approx 400$ ps in (Sb$_{1-x}$Bi$_x$)$_2$Te$_3$, where the chemical potential is positioned inside the bulk insulating gap.[64] In particular, a recent time-resolved ARPES study provided evidence of the population inversion in the p-type 3D TI (Sb$_{0.7}$Bi$_{0.3}$)$_2$Te$_3$ on the timescale of 10 ps,[65] which is two orders of magnitude larger than in graphene. The prolonged lifetime of population inversion in 3D TIs with chemical potential close to the Dirac node is attributed to the relaxation bottleneck originating from the reduced phase space near the node.

It is still an open question whether population inversion can be realized in 3D DMs. Several experimental reports suggested that the ultrafast carrier dynamics in 3D DMs is qualitatively similar to that of graphene. The overall lifetime of photoexcited carriers was found to be of the order of a few picoseconds.[59,60] A recent study by Zhang et al.[61] showed evidence of population inversion in Cd$_3$As$_2$ DSM on the timescale of 6 ps. This finding suggests that prolonged population inversion can be established at least in some 3D DMs. The possibility of achieving an inverted population of electrons and holes by optical pumping, makes optically excited DMs a promising system for realizing a transient excitonic instability.[15,16]

![Figure 1](image_url)

**Figure 1.** Transient excitonic instability is a pumped DM with a population inversion. Before the pump, at time $t < t_0$, the system is in equilibrium and is described by a single chemical potential $\mu = 0$; at $t = t_0$ electrons are excited from the valence band to the conduction band; after thermalization time $t = t_1$, a population inversion is established, that is, photoexcited electrons and holes form two distinct Fermi-Dirac distributions with chemical potentials $\mu_e$ and $\mu_h$, respectively; pairing between finite populations of electrons and holes leads to formation of a transient excitonic state at $t = t_2$, characterized by gap openings at non-equilibrium chemical potentials; due to a finite lifetime of the population inversion, transient excitonic states decay towards equilibrium ($t = t_3$). Panels on the right show schematically the relaxation of the chemical potentials $\mu_e$ and $\mu_h$, and the order parameter $\Delta$ toward equilibrium. For illustration, we consider a single Dirac cone and only show the 2D energy dispersion of the Dirac states (for a 3D DM, we assume a 2D projection of the 3D Dirac states). Empty Dirac states are indicated in white, filled states in yellow.

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By definition, excitonic instability in equilibrium occurs when the exciton binding energy exceeds the (positive or negative) band gap of the material. This can be realized in narrow gap semiconductors, semimetals, or metals with overlapping bands. In these systems, at sufficiently low energies, the Coulomb attraction between electrons and holes residing in the conduction and valence band, respectively, leads to a new collective ground state known as an excitonic insulator, or the electron-hole Bardeen–Copperschrieffer (BEC) state, that is, a condensate of electron–hole Cooper pairs.\cite{65,66,67} This should be distinguished from a Bose–Einstein condensate (BEC) of excitons, or bound states of a single electron–hole pair.\cite{68,69} A transition from an exciton BEC to a BCS state has been predicted for increasing densities of electrons and holes at low temperatures.\cite{70}

Many systems have been suggested as suitable for excitonic condensation. These include systems of "direct" excitons, for example, narrow-gap semiconductors where paired electrons and holes reside in the conduction and valence bands of the same material.\cite{66,67} Signatures of the excitonic insulator phase have been reported in semiconductors $\text{TmSe}_{0.45}\text{Te}_{0.55}$,\cite{71} $\text{TaS}_2$,\cite{72} and $17\text{-T},\text{Se}_2$.\cite{73} Another example is condensation of "indirect" excitons realizable in bilayer systems, where electrons and holes are spatially separated.\cite{74,75} Evidence of BEC of indirect excitons has been reported in electron-hole bilayers found in semiconductor quantum wells.\cite{79,80,81} However, the most clear signatures of the exciton BEC have been observed in quantum Hall electron–electron bilayers in strong magnetic field.\cite{83,84} Gated graphene bilayers\cite{87,88} and TI thin films\cite{90,91} have also been proposed as promising platforms. Recently, a bilayer exciton BEC has been proposed in quasi-one-dimensional systems with a single particle hybridization between electron and hole populations.\cite{92}

In principle, electron–hole pair formation is also possible in highly excited semiconductors, where electron and hole populations are created by optical pumping in the conduction and valence bands respectively. In this case the possible excitonic insulator phase or the exciton BEC are intrinsically non-equilibrium phenomena due to finite lifetime of electron and hole populations. As commonly accepted in the literature, in this work we refer to these states collectively as a transient excitonic condensate. Such systems have been studied extensively in theoretical works.\cite{17,19,93,94} Experimentally, clear signatures of a transient excitonic condensate are yet to be observed. There are indications of possible ordered exciton state in $\text{CuO}$,\cite{95} Also, signs of preformed (uncondensed) electron–hole pairs in measurements of stimulated emission in highly excited $\text{ZnO}$ were reported.\cite{18}

More recently, pumped black phosphorus was suggested as a promising candidate for excitonic condensation, with lifetime of carriers excited across the band gap exceeding 400 ps.\cite{100}

In a different setting, semiconductor-based microcavities provide a convenient platform for condensation. In these systems photons are confined and strongly coupled to electronic excitations, leading to the creation of exciton polaritons. Condensation is favorable due to the very small mass of the polaritons and BEC of exciton polaritons has indeed been observed in these systems.\cite{101,102,103} Among other work on non-equilibrium many-body states, we mention Floquet exciton condensation in bilayer graphene\cite{104} and photo-induced superconducting states in semiconductors under optical driving.\cite{105}

The proposal for realization of the transient excitonic condensate in optically-excited DMs differs from previous work in several important aspects. First, we consider a semimetal with a very special energy dispersion. The linear (Dirac) dispersion relation in DMs results in the strongly energy-dependent density of states (DOS): $\mathcal{N}(E) \propto E$ or $\mathcal{N}(E) \propto E^2$ in 2D and 3D DMs, respectively. The presence of nodes in the low-energy spectrum implies vanishing DOS at the nodes, which leads to a critical coupling for many-body instabilities.\cite{106}

The strength of electron–electron interactions is typically characterized by a material-specific parameter, the so-called dimensionless coupling constant $\alpha$, which is expressed as a ratio between the Coulomb energy and the kinetic energy. In the case of DMs, it is given by $\alpha = e^2/hv$, where $e$ is the material-dependent dielectric constant and $v$ is the Fermi velocity of the linearly dispersing states. Critical coupling, $\alpha_c$, is defined as a value of the dimensionless coupling constant such that for $\alpha < \alpha_c$, the system is not significantly affected by interactions and the spectrum remains gapless, while for $\alpha > \alpha_c$, the spectrum acquires an energy gap due to interactions. The gapped phase can be referred to as the excitonic insulator. Many theoretical studies suggest that in the case of 2D DM (graphene), $\alpha_c \approx 1$.\cite{106,107,108,109} The excitonic insulator phase has not yet been observed in DMs. Experimental studies on suspended graphene, where $\alpha$ is expected to be larger than 1 ($\alpha \approx 2$), show that the only effect of electron–electron interactions is the logarithmic renormalization of the Fermi velocity close to the node.\cite{110}

Optical pumping can be used to generate excitonic gaps in DMs. Second, optical pumping in DMs allows for a tunable enhancement of the DOS of the inverted electron and hole populations, and, thus, offers a tuning knob for the effective Coulomb interaction. This is a unique feature of DMs, which arises from the strongly energy-dependent DOS. For instance, it is not achievable in metals or semiconductors in 2D, since in this case the DOS is constant at low energies. Finally, an important signature of the transient excitonic state are the energy gaps that open up in the quasiparticle spectrum at the non-equilibrium chemical potentials for electrons and holes. Although the gaps have a transient nature, our proposal, if realized, offers an alternative way of generating band gaps in DMs, which is highly desirable for application in optoelectronics.

Here, we develop a unified theoretical description of transient excitonic states in optically pumped DMs, based on our previous work on 2D\cite{13} and 3D DMs.\cite{16} We analyze the predictions of the model for specific materials within this common framework. The theory is based on a low-energy effective model for DMs. As a first step, we consider excitonic instabilities in 2D and 3D DMs with population inversion by using the mean-field BCS theory, applied to a system with two types of carriers (electrons and holes). Metallic screening is important in pumped DMs, where the non-equilibrium chemical potentials are located far from the Dirac node. Screening effects are included in the Thomas–Fermi approximation. By substituting material parameters in the model, we obtain numerical estimates for the values of critical temperature ($T_c$) and excitonic gap ($\Delta$) for realistic materials. The relaxation of the transient excitonic state towards equilibrium is studied using a dynamical model based on semiconductor Bloch equations.
The largest effect, that is, a gap of the order of 10 meV and a critical temperature of 70 K, is expected for pristine suspended graphene, in which pumping is realized selectively on a single valley. Large gaps and critical temperatures should enable detection of the transient excitonic states in graphene by trARPES; however, the relatively short lifetime (100 – 200 fs) is likely to make the observation challenging. Another promising 2D system is a 3D TI with a single surface Dirac cone such as Bi$_2$Se$_3$ and related materials. Although the estimated values for the gap and $T_c$ are typically smaller in 3D TIs compared to graphene due to smaller $\alpha$, the lifetime of the population inversion is at least an order of magnitude larger. Further tuning material parameters such as the bulk gap and the dimensionless coupling constant, can lead to a much larger effect in 3D TIs, namely $T_c$ of the order of 100 K and gaps of 10s of meV. In the case of DSM and WSM, severe screening effects due to large Dirac cone degeneracy in existing materials, limits the estimated gaps sizes to 1 meV and critical temperature to few K. By using the recipe for enhancement of the excitonic gap and $T_c$, similarly to the case of 2D DMs, large values, comparable to those predicted for graphene, could be also obtained in future 3D DMs.

The rest of the paper is organized as follows. In Section 2, we present the mean-field theory of excitonic pairing in pumped DMs. In Section 3, we discuss spectroscopic signatures and calculate the order parameter as a function of temperature, non-equilibrium chemical potentials and dimensionless coupling constant. Furthermore, we define clear criteria for the efficiency of optical pumping in generating transient excitonic states. In Section 4, we summarize the numerical estimates for several examples of realistic and hypothetical 2D and 3D DMs and suggest experimental setups in which the effect could be observed. We also derive general criteria for achieving large excitonic gaps and $T_c$ and suggest routes for search of promising material candidates. Finally, in Section 5 we offer some conclusions and outlook.

2. General Theory of Pumped DMs

2.1. Pumping Scheme

Pump-probe experiments suggest that population inversion can be achieved in optically excited DMs. The pumping scheme may differ depending on the material. In graphene, Dirac states extend in the energy window of approximately 1 eV; therefore, an optical excitation promotes electrons from the occupied states in the lower Dirac cone into the unoccupied states in the upper Dirac cone (this situation is depicted in Figure 1). In a typical 3D TI like Bi$_2$Se$_3$, with a band gap of few hundred meV, a pump pulse excites electrons from the bulk valence band into the bulk conduction band. The excited carriers then cascade into the unoccupied states in the upper Dirac cone. Vanishing phase-space for excitations near at the Dirac point acts as a bottleneck for relaxation processes. Pumping on 3D DMs occurs in a similar fashion since the energy extent of the 3D Dirac states in typically small compared to the energy of the pump pulse. However, recently dynamics of photoexcited carriers was probed in 3D DMs by direct excitation with low-energy photons\cite{60} similarly to graphene.

Based on these observations, a schematic diagram of a pumped DM is drawn in Figure 1.

In equilibrium ($t < t_0$), we consider a DM with a chemical potential at the Dirac point ($\mu = 0$). The pump pulse is switched on at $t = t_0$ and promotes electrons from the occupied states below to the empty states above the Dirac point. We assume that after rapid thermalization, the population inversion is established at $t = t_1$ and relaxes towards a single Fermi-Dirac distribution on the timescale $t_1$ (in general, at a higher temperature than in equilibrium). The lifetime is of the order of 100 fs in graphene\cite{46} however, we should note that there are other trARPES reports that do not show any population inversion in graphene within the temporal resolution of the experiment\cite{63}. In 3D TIs, the lifetime is extended to at least several ps in recent reports\cite{63}. Suppression of Auger scattering that was theoretically predicted to occur in WSMs, could lead to population inversion with similar lifetimes\cite{111}.  

2.2. Model Hamiltonian with Interactions

For the discussion presented in this section, we assume that the lifetime of the population inversion, established at $t = t_1$ (see Figure 1), is infinitely long so that the system can be considered to be in quasiequilibrium with chemical potentials $\mu_{e/h}$, for electron/hole pockets. We will then derive a general Hamiltonian for a pumped DM including Coulomb interactions.

The pumped system in the quasiequilibrium state can be described by the Hamiltonian

$$H = H_e + H_h + \mathcal{V}'$$

(1)

where the first two terms represent the non-interacting Hamiltonians for electrons and holes, respectively, and the last term is the electron-hole interaction, written in the band basis. The Hamiltonian in Equation (1) can be formally derived by diagonalizing the interacting Dirac Hamiltonian $H$ for a 2D or 3D DM and by including the finite chemical potential for electron and hole bands (the procedure is demonstrated in ref. [16] for the case of a DSM/WSM). Thus the electron and hole Hamiltonians are essentially the linearly-dispersing conduction and valence band of the DM, modified by optical pumping

$$H_{e/h} = \sum_{\alpha} \sum_{k} \epsilon_{k} \sigma_{\alpha} \epsilon_{k}^{e/h} \sigma_{\alpha}$$

(2)

where $\epsilon_{k}^{e/h} = \pm \hbar c k - \mu_{e/h}$ is the dispersion for the electron/hole populations, $\mu_{e/h}$ is the electron/hole chemical potential, and $\nu$ is the Fermi velocity of the Dirac states. Here we allow for multiple Dirac cones labeled by index $\alpha$, for example, $\alpha = 1, 2$ in graphene or $\alpha = 1, 2N$, where $N$ is an integer, in a WSM; $\sigma_{\alpha}^{x}$ ($\sigma_{\alpha}^{y}$) is the creation(annihilation) operator for an electronic state with momentum $k$ in the band $\sigma = \{e, h\}$ belonging to the Dirac cone $\alpha$. Equations (1) and (2) can be used to study 2D or 3D DMs by letting $\mathcal{V}$ be a 2D or 3D momentum, respectively. The original particle–particle interaction term written in the spinor basis is given by

$$V = \sum_{\alpha, \beta, \alpha', \beta'} \sum_{k, q, \sigma} \sum_{\alpha, \beta, \alpha', \beta'} \Phi_{\bar{k} + q, \sigma}^{\alpha'} \Phi_{\bar{k}, \sigma}^{\alpha} \Phi_{k, \sigma}^{\beta'} \Phi_{k, \sigma}^{\beta}$$

(3)
where $\Phi_{a}^{\nu}$ is the spinor corresponding to a state with momentum $K$ and spin $\sigma$ in a Dirac cone $\alpha$. One obtains the interaction term $\mathcal{V}$ in the band basis by expressing the spinors in the diagonal, or band, basis as $\Phi_{q.a}^{\nu} = \sum_{\nu' = \nu} \Phi_{q.a}^{\nu' \nu}$, where $\Phi_{q.a}^{\nu' \nu}$ and $\chi_{q.a}^{\nu' \nu}$ are the eigenvectors of the Dirac Hamiltonian $H$. The expression for $\mathcal{V}$ depends on the form of $H$ considered and will, in general, inherit the spinor structure of the Dirac Hamiltonian.

At the simplest level, considering a spinless model of the DM and only intra-nodal interactions, that is, interactions between holes and electrons belonging to the same Dirac cone, the interaction term can be written as

$$\mathcal{V} = \sum_{q} \sum_{q'} \sum_{\sigma, \sigma'} \mathcal{V}_{\sigma}(q, q') \sum_{\alpha, \beta} \phi_{\alpha}^{\sigma} \phi_{\beta}^{\sigma'} \phi_{\alpha'}^{\sigma'} \phi_{\beta'}^{\sigma}$$

where $\mathcal{V}_{\sigma}(q, q')$ is the screened Coulomb potential. In this case, the number and degeneracy of the Dirac cones will be taken into account in the form of $\mathcal{V}_{\sigma}(q)$. This simplified model was used in ref. [15] to describe the transient excitonic insulator phase in 2D DMs.

A more general form of the interaction potential can be derived by considering intra- and inter-nodal excitonic pairing of the form $\Phi_{q.a}^{\nu} \Phi_{q'.a'}^{\nu'}$. For a WSM with broken time reversal symmetry, that is, a 3D DM with two non-degenerate Dirac nodes ($q = 2$), we adopt the notations $q \rightarrow q$, where $q = k \mp K$, where $\pm K$ is the position of the two nodes with different chirality. The interaction potential for this case in the band basis is given by

$$\mathcal{V} = \sum_{q \neq q'} \sum_{\sigma, \sigma'} \mathcal{V}_{\sigma}(q, q') \sum_{\alpha, \beta} \phi_{\alpha}^{\sigma} \phi_{\beta}^{\sigma'} \phi_{\alpha'}^{\sigma'} \phi_{\beta'}^{\sigma}$$

$$+ \mathcal{V}_{\sigma}(q, q') \sum_{\alpha, \beta} \phi_{\alpha}^{\sigma} \phi_{\beta}^{\sigma} \phi_{\alpha}^{\sigma'} \phi_{\beta}^{\sigma'}$$

where $\mathcal{V}_{\text{intra}}$ and $\mathcal{V}_{\text{inter}}$ are the intra- and inter-nodal interaction potentials, respectively,

$$\mathcal{V}_{\text{intra}}(q, q') = \mathcal{V}_{\text{intra}}(q - q') \left[ \frac{\sin \theta \sin \theta'}{2} + \frac{1 + \cos \theta \cos \theta'}{2} \times \cos(\phi - \phi') \right]$$

$$\mathcal{V}_{\text{inter}}(q, q') = -[2V_{s}(2K) - V_{s}(q - q')(1 + q \cdot q')]$$

Here, $[q, \theta, \phi]$ are the polar coordinates and $2K$ is the separation between the nodes. Note that in Equations (7) and (8) we only kept the leading terms in the interaction potential by using $|q - q'| << |q - q' - 2K|$ and $|q - q'| << |K|$.

In the next section, we will discuss the form of the screened Coulomb potential $\mathcal{V}_{\sigma}(q)$ for a pumped DM.

### 2.3. Screened Coulomb Interaction

In the static random phase approximation, the dielectric function $\varepsilon(q, \omega = 0)$, the screened Coulomb potential $\mathcal{V}_{\sigma}(q)$, and the screening wavevector $\kappa$ are given by ref. [112].

$$\varepsilon(q, 0) = \begin{cases} 1 + \frac{\kappa}{q} & 2D \\ 1 + \frac{\kappa^2}{q^2} & 3D \end{cases}$$

$$\mathcal{V}_{\sigma}(q) = \begin{cases} \frac{V_{\text{DM}}(q)}{\varepsilon(q, 0)} = \frac{2\pi c^2}{\varepsilon q + q'} & 2D \\ \frac{V_{\text{inter}}(q)}{\varepsilon(q, 0)} = \frac{2\pi c^2}{\varepsilon q + q' + \kappa^2} & 3D \end{cases}$$

where we have explicitly specified the expressions for a 2D and 3D system. In Equation (10), $\varepsilon$ is the dielectric constant of the material and $V_{\text{2D}}(q) = \frac{2\pi c^2}{\varepsilon q + q'}$ is the unscreened Coulomb potential in 2D(3D), which is the Fourier transform of the real space Coulomb potential $V(r) = \frac{1}{r}$.

A system with population inversion consists of electron and hole pockets, which are in general characterized by different densities $n_i$ and chemical potentials $\mu_i$, $i = e, h$. Hence, in Equation (11), we defined the global screening wavevector, which combines electron and hole contributions to screening. Alternatively, $\kappa$ can be expressed directly in terms of the screening wavevectors for electrons and holes.

$$\kappa = \kappa_e + \kappa_h$$

$$\kappa^2 = \kappa_e^2 + \kappa_h^2$$

We will now derive the expressions for $\kappa_i$, $i = e, h$ in the case of DM, that is, for a system with linear dispersion $E = \hbar v k$. We assume a particle-hole symmetric spectrum with identical velocities for electrons and holes, and use the Thomas–Fermi approximation, that is, $T \rightarrow 0$, in the expression for the screening wavevector. The Fermi wavevector is given by $k_{F} = \mu_{i} / \hbar v$. In the case of 2D DM, the density is $n_{i} = N_{i} / A = \frac{\sqrt{2} \xi}{\sqrt{A}}$, where $N_{i}$ is the number of quantum states, $A$ is the area, and $g$ is the degeneracy. In the case of 3D DM, $n_{i} = N_{i} / V = \frac{\sqrt{2} \xi}{\sqrt{V}}$, where $V$ is the system volume. Using the definition of the screening wavevector in 2D and 3D, given in Equations (12) and (13), we get

$$\kappa_{i} = \begin{cases} \frac{g c^2}{\hbar} \mu_{i} \equiv g a k_{i}^{2} / \hbar & 2DDM \\ \sqrt{\frac{2g c^2 v h}{\pi e}} \mu_{i} \equiv \sqrt{\frac{2g c^2 v h}{\pi e}} k_{i}^{2} / \hbar & 3DDM \end{cases}$$
2.4. Gap Equation

2.4.1. General Derivation

Having established the form of the interacting Hamiltonian, we will now proceed with the derivation of the self-consistent equation for the mean-field order parameter, or excitonic gap. In order to obtain the gap equation in the general form, we write down the Hamiltonian of a pumped DM [Equation (1)], including the interaction term in the band basis

\[ \mathcal{H} = \sum_{\Delta q \neq 0} E_{\Delta q} c_{\Delta q}^\dagger c_{\Delta q} + \sum_{q \neq 0} \tilde{V}(q, \Delta q) c_{q}^\dagger c_{\Delta q} + \sum_{q \neq 0} \tilde{V}(q, \Delta q) c_{\Delta q}^\dagger c_{q} \]  

(15)

We consider pairing between electrons and holes in 2D or 3D momentum space, with a general interaction potential \( \tilde{V}(q, \Delta q) \). For simplicity we have omitted the node subscript \( \alpha \). The interaction term in Equation (15) can be easily adjusted to represent intranodal or internodal interactions for a general system with multiple nodes. More specifically, for intranodal interactions, \( c_{q}^\dagger c_{\Delta q} \) is the creation(annihilation) operator for the band \( \tau = e, h \) of the same node \( \alpha \). For internodal interactions, \( c_{q}^\dagger c_{\Delta q} \) and \( c_{\Delta q}^\dagger c_{q} \) are, respectively, the creation(annihilation) operators for the conduction and valence bands belonging to different nodes; also note that momentum \( q \) refers to a specific node, \( q \equiv k + K \), where \( \pm K \) is the position of the node. The interaction potential \( \tilde{V}(q, \Delta q) \) takes on the form derived in Section 2.2 for intra- or internodal pairing. Equations (7) and (8).

Furthermore, we introduce the electron(hole) Green’s functions and the anomalous Green’s function.

\[ G_{e(h)}(q, t - t') = -< T_{c_{q}c_{q}^\dagger}(t)c_{q}c_{q}^\dagger(t') > \]  

(16)

\[ F(q, t - t') = -< T_{c_{q}c_{q}^\dagger}(t)c_{q}c_{q}^\dagger(t') > \]  

(17)

where \( T_{t} \) is the imaginary time-ordering operator. In the mean-field approximation, the order parameter, or excitonic gap \( \Delta(q) \), is calculated as

\[ \Delta(q) = \sum_{q'} \tilde{V}(q, q') \langle c_{q'}^\dagger c_{q'} \rangle \]  

\[ = T \sum_{q', i \omega_n} \tilde{V}(q, q') F(q'; i \omega_n) \]  

(18)

where \( \omega_n = (2n + 1)\pi / \beta \) is a fermionic Matsubara frequency, \( \beta = 1/k_B T \), \( k_B \) is the Boltzmann constant, and \( T \) is the temperature assumed to be the same for photoexcited electrons and holes. In order to obtain the gap equation, one can use, for example, the Gor’kov approach, which is based on time-dependent equations of motions for \( G_{e(h)} \) and \( F_{e(h)} \). The final result for the gap equation reads

\[ \Delta(q) = \sum_{q'} \tilde{V}(q, q') \frac{\Delta(q')}{\omega_q(q') - \omega_n(q')} \left[ n_T(\omega_q) - n_T(\omega_q) \right] \]  

(19)

where

\[ \omega_q(q) = \frac{E_0^q + E_h^q}{2} \pm \frac{1}{2} \sqrt{\left(E_0^q - E_h^q\right)^2 + 4|\Delta(q)|^2} \]  

(20)

are the renormalized excitonic bands and \( n_T(\omega_q) = 1/(e^{\omega_q/k_BT} + 1) \) is the Fermi-Dirac distribution. In equilibrium, i.e. in the limit \( \mu_c = -\mu_h = 0 \), one recovers the usual BCS gap equation.

2.4.2. Intranodal versus Internodal Interactions

Using the results of the previous section, we will discuss the competing excitonic phases originating from internodal and intranodal interactions for the specific case of 3D DMs. Note that here we consider either a DSM or a WSM with broken time reversal symmetry. Taking into account different interactions present in the system, we can specify the form of the general order parameter introduced in Equation (18)

\[ \Delta(q) = \sum_{q'} \tilde{V}(q, q') \langle c_{q'}^\dagger c_{q'} \rangle \]  

(21)

For \( \alpha = \beta \), \( \Delta(q) \) describes pairing between electrons and holes in the same node (intranodal interactions), while for \( \alpha \neq \beta \), pairing occurs between electrons and holes belonging to different nodes (internodal interactions). Intranodal pairing leads to an excitonic insulator (EI) phase. Internodal pairing gives rise to a charge density wave (CDW) phase with the modulation momentum equal to the distance \( 2K \) between the nodes. Equilibrium excitonic phases in a WSM with broken time reversal symmetry
were studied in refs. [117–119]. In the case of short-range (con- tact) interaction potential, the EI phase is more energetically fa-
vorable. In the case of unscreened Coulomb potential, the CDW phase becomes dominant. We will show in Section 3.2 that this result holds in the case of the screened Coulomb potential in a pumped WSM, with an important difference that both the EI phase and the CDW phase are present at arbitrarily weak cou-
pling.

We will further specify the form of the order parameter and the gap equation for intranodal and internodal interactions in a pumped system. The internodal part of the interaction potential was introduced in Equation (7). Keeping only the slowest vary-
ing term in the angular-dependent part of the potential proportion-
to \cos(\phi - \phi')/2, we get \( V_{\text{intra}} = V'(q - q') \cos(\phi - \phi')/2 \). The self-consistent gap equation becomes

\[
\Delta^\alpha(q) = \sum_{q'} 2 V_{\text{intra}}(q', q) \frac{\Delta(q')}{\omega_+^\alpha(q') - \omega_-(q')} \left[ n_\uparrow(\omega_+) - n_\downarrow(\omega_-) \right] - \frac{1}{(2\pi)^3} \int \frac{d^3q'}{V'(q - q') \Delta^\alpha(q') \cos(\phi - \phi')} \times \left[ n_\uparrow(\omega_+) - n_\downarrow(\omega_-) \right] dV
\]

and the excitonic gap is given by

\[
\Delta^\alpha(q) = \sum_{q'} V_{\text{intra}}(q, q') \left( \epsilon_{q', \tau} \epsilon_{q, \tau}^\dagger \right), \quad \alpha = 1, 2
\]

The mean-field Hamiltonian of the system with intranodal interac-
tions reads

\[
H_{\text{intra}} = \sum_{q, \alpha, \tau} \epsilon^\alpha_q c_{q, \tau}^\dagger c_{q, \tau} - \sum_{q, \alpha} \Delta^\alpha(q) c_{q, \tau}^\dagger c_{q, \tau} - \sum_{q, \alpha} \tilde{\Delta}^\alpha(q) c_{\tau}^\dagger c_{\tau} - \sum_{q, \alpha} \Delta^\alpha(q) c_{q, \tau}^\dagger c_{q, \tau}^\dagger
\]

where \( \Delta^\alpha(q) = 2 \tilde{\Delta}^\alpha(q) \). The first term in Equation (24) is the non-
interacting Hamiltonian of the two nodes while the last two terms describe interactions within each node.

The internodal part of the interaction potential is given by Equation (8). Furthermore, by neglecting the angular-dependent part proportional to \( q - q' \), the interaction potential is simply the screened Coulomb potential \( V_{\text{intra}}(q - q') \). In this case, the self-
consistent gap equation reads

\[
\Delta^\prime(q) = \sum_{q'} V_{\text{inter}}(q, q') \frac{\Delta(q')}{\omega_+^\prime(q') - \omega_-(q')} \left[ n_\uparrow(\omega_+) - n_\downarrow(\omega_-) \right] - \frac{1}{(2\pi)^3} \int \frac{d^3q'}{V'(q - q') \Delta^\prime(q') \cos(\phi - \phi')} \times \left[ n_\uparrow(\omega_+) - n_\downarrow(\omega_-) \right] dV
\]

and the excitonic gap is given by

\[
\Delta^\prime(q) = \sum_{q'} V_{\text{inter}}(q, q') \left( \epsilon_{q', \tau} \epsilon_{q, \tau}^\dagger \right), \quad \tau = e, h, \quad \tau' \neq \tau
\]

The corresponding mean-field Hamiltonian reads

\[
H_{\text{inter}} = \sum_{q, \alpha, \tau} \epsilon^\alpha_q c_{q, \tau}^\dagger c_{q, \tau} - \sum_{q, \alpha} \Delta^\prime(q) c_{q, \tau}^\dagger c_{q, \tau}^\dagger - \sum_{q, \alpha} \tilde{\Delta}^\prime(q) c_{\tau}^\dagger c_{\tau}^\dagger
\]

As in Equation (24), the first term in Equation (27) is the non-
interacting part of the Hamiltonian, while the last two terms represent interactions between the nodes.

The CDW gap equation, Equation (23), does not contain any explicit angular dependence (apart from the screened Coulomb potential \( V_{\text{intra}}(q - q') \), which depends on \( q - q' \)); hence the corresponding order parameter is isotropic. In contrast to this, the EI gap equation, Equation (22), contains a factor \( \cos(\phi - \phi') \). By taking \( \cos(\phi - \phi') = (e^{i(\phi - \phi')} + e^{i(\phi' - \phi)})/2 \) and re-defining the EI gap as \( \Delta^\alpha(q) = \Delta^\alpha(q) \cdot e^{i\phi} \), one obtains a self-consistent equation for the magnitude of the EI gap \( \Delta^\alpha(q) \), which is identical to the CDW gap equation with an additional factor of 1/2. Hence, the EI gap is always smaller than the CDW gap for the same model parameters. (One exception is the special (hypothetical) case \( g = 1 \), for which only one node has population inversion and therefore only intranodal interactions leading to the EI phase are relevant). Furthermore, the screened Coulomb potential in the gap equation can be replaced by its angle average, \( V_{\text{intra}}(q - q') \rightarrow \left\langle V_{\text{intra}}(q, q') \right\rangle \), which depends only on the magnitudes of vectors \( q \) and \( q' \).

Since the order parameter is momentum-dependent, we will solve the self-consistent gap equation numerically on a finite momentum mesh. We will use the dimensionless units \( \Delta \rightarrow \Delta/hv_\lambda, q \rightarrow q/\lambda, T \rightarrow k_B T/hv_\lambda \), where \( \lambda \) is the momentum cut-off of the Dirac Hamiltonian in Equation (2). The corresponding cutoff energy scale is \( \Lambda \equiv hv_\lambda \). From the solutions of the gap equation at zero and finite temperatures, we will construct the phase diagrams of the transient excitonic condensate and calculate the observable quantities such as the spectral function and DOS.

### 3. Properties of Transient Excitonic Condensate

Assuming a total number of nodes \( N \) in the system, one can consider the following cases for optical pumping: i) uniform pumping, that is, all nodes are pumped and population inversion is realized for each node, and ii) selective pumping, that is, population inversion is realized for a certain number of nodes \( N_g \leq N \). For \( N > 1 \), pairing within a single node is affected by screening of carriers belonging to other nodes, provided there is a finite density of electrons and holes at the node. In the expression for the Thomas–Fermi screening wavevector (see Table 1), the degeneracy fac-
tor \( g \) essentially represents the number of cones which con-
tribute to screening. Importantly, screening becomes stronger for larger \( g \).

For a Bi₈Se₃-type 3D TI with a single Dirac node on the sur-
face (\( N = 1 \)), we have \( g = 1 \). For a WSM with two non-degenerate nodes (\( N = 2 \)), case i) corresponds to \( g = 2 \), while case ii) corre-
sponds to \( g = 1 \) in the calculation of the screening wavevector.

In graphene, there is an additional spin degeneracy of the Dirac
states and therefore case i) corresponds to $g = 4$, while case ii) corresponds to $g = 2$ (valley-selective pumping). 3D DMs can have a large number of nodes, for example, $N = 24$ in TaAs WSM. In such systems, if all nodes are uniformly pumped, excitonic effects will be inhibited by screening.

Furthermore, for systems with $N > 1$, it is important to clarify which type of interactions is relevant in the two pumping cases. For the simplest case of a WSM with two nodes and for uniform pumping ($g = 2$), both intranodal and internodal interactions are present and therefore both the EI and CDW phases are realized. For selective pumping on a single Weyl node ($g = 1$), one of the nodes has a population inversion, while the other one is in equilibrium. Throughout this work, we assume that the equilibrium chemical potential is located at the node, leading to vanishing DOS. In this case, the strongest pairing is realized for intranodal interactions within the pumped node, leading to the EI phase. The CDW gap rapidly vanishes as a function of the mismatch between the equilibrium and non-equilibrium chemical potentials. Similarly, the EI gap vanishes for $\mu_e \neq -\mu_h$.\[15\]

For a DSM with the minimal degeneracy $g = 2$, only intranodal interactions are included. For a DSM with $g \geq 4$, both intranodal and internodal interactions are relevant. Importantly, in all cases where internodal interactions are important, for example, for $g > 1$, the CDW gap is always larger than the EI gap. Therefore, we expect that in pumped 3D DMs under realistic experimental conditions the transient excitonic state is manifested in a CDW phase.

### 3.1. Spectroscopic Signatures

In the pumped DM, the excitonic phase is characterized by energy gaps that open up at the non-equilibrium chemical potentials (Figure 2). In the DOS and the spectral function, the gaps separate filled states below from the empty states above the chemical potentials. The vanishing DOS in the gaps could be detected by scanning tunneling microscopy. The spectral function can be probed by trARPES. A feature that could be directly detected in the ARPES spectra is the bending and vanishing of the spectral weight of the occupied bands near the non-equilibrium chemical potentials.

The excitonic gap in a pumped DM is a dynamical quantity due to the transient nature of the photoexcited Dirac states. As the electron and hole chemical potentials approach the equilibrium value ($\mu = 0$ before pumping) over the lifetime of the population inversion, the position of the gap will move towards the node and the size of the gap will decrease. This is shown schematically in Figure 1. A more detailed discussion of the timescales involved
and the relaxation of the order parameter based on rate equation is presented in Section 4.3.

### 3.2. Tunability of the Critical Coupling

In equilibrium DM, there is a critical value \( a^0 \) of the dimensionless coupling constant such that for \( a > a^0 \) the system becomes an excitonic insulator.\(^{[106]}\) In this section, we will discuss the reduction of the critical coupling which is expected in pumped DMs due to the enhanced DOS away from the Dirac node.

In the case of 2D DM, analytical analysis of the gap equation has been carried out in the limit of strong screening, when the Coulomb potential becomes a contact interaction in position space or, equivalently, a constant in momentum space \( V(q) \rightarrow V_0 = 2\pi n a / \kappa \),\(^{[10]}\) where \( \kappa \) is the screening wavevector. The important result of the analytical model is that for perfectly matched chemical potentials (\( \mu_c = -\mu_b \)), the excitonic insulator phase is realized for any finite value of \( \mu_b / h \). The calculated excitonic gap \( \Delta \) and the critical temperature \( T_c \) increase monotonically with increasing \( \mu_b / h \).

In the case of the realistic screened Coulomb potential, the phase diagram for the order parameter is more complex due to the competition between screening and the enhanced DOS for a given \( \mu_b / h \). As a result of this competition, pumping is more advantageous for excitonic condensation compared to equilibrium only in a certain range of the parameter space defined by material parameters, \( a, \mu_b, \) and \( g \). This crucial point is demonstrated below for 2D and 3D DMs.

We consider pumping to be efficient if i) for \( a < a^0 \), the transient excitonic gap is different from zero, or ii) for \( a > a^0 \), the transient excitonic gap is larger than the equilibrium gap. In what follows we introduce for convenience a single quasi-equilibrium chemical potential \( \bar{\mu} = \mu_c = -\mu_b \). In particle-hole symmetric systems, population inversion with balanced chemical potentials can be realized if the chemical potential is positioned at the Dirac point before pumping. Note that this condition may differ in DMs which have different Fermi velocities for electrons and for holes.

**Figure 3** shows the phase diagram of the transient excitonic state in the \( \bar{\mu} - a \) plane for a 2D DM. Here we consider the regime \( a \lesssim a^{1D} \), where \( a^{1D} \approx 1.0 \) is the equilibrium critical coupling in 2D. In equilibrium, the Coulomb potential becomes unscreened when \( \mu = 0 \). In this case, for numerical calculations with the Thomas–Fermi model we use a small but finite value of the screening wavevector such that variations around this value do not change appreciably the result for \( a^0 \). The critical coupling in a pumped system is defined for a given \( \bar{\mu} \) as the minimum value of \( a \) for which the gap is different from zero. In practice, we use a condition \( \Delta_{\text{max}} \geq \delta \), where \( \Delta_{\text{max}} \) is the maximum value of the gap and \( \delta \) is a small number (\( \delta = 10^{-6} \) in units of energy). In Figures 3 and 4a, the critical coupling defines a line separating the dark (\( \Delta_{\text{max}} = 0 \)) and bright (\( \Delta_{\text{max}} \neq 0 \)) regions of the phase diagram. As one can see from Figure 3, the critical coupling is significantly reduced in the pumped state, where \( \bar{\mu} \neq 0 \). A similar behavior is found for a 3D DM, as demonstrated in Figure 4a for a WSM with \( g = 1 \) and for \( \alpha \) in the range \( 0 \leq \alpha \leq 3 \).

The tunability of the critical coupling with pumping is further confirmed in Figure 4b and **Figure 5** which show the scans of the \( \alpha - \bar{\mu} \) phase diagram (\( \Delta_{\text{max}}(\alpha) \) curves), for a 3D DM with \( g = 1, 2, 3 \), and for increasing values of the chemical potential \( \bar{\mu} \). In equilibrium (\( \bar{\mu} = 0 \)), the values of the critical coupling for the CDW and the EI phases are given by \( a_{\text{CDW}} \approx 3.0 \) and \( a_{\text{EI}} \approx 1.5 \), respectively, in agreement with the values obtained analytically in ref. [119].

In the pumped case (\( \bar{\mu} \neq 0 \)), the critical coupling for excitonic instability vanishes. In the range \( \alpha \leq a_{\text{CDW(\bar{\mu})}} \), the equilibrium system remains gapless, while the pumped system develops a
3.3. Phase Diagrams of the Excitonic Condensate

Figure 6 illustrates the effect of pumping on the $T - \bar{\mu}$ phase diagrams. The critical temperature $T_c$ is defined as the minimum value of $T$ for which the excitonic gap is different from zero. In Figure 6, the following cases are considered: i) a 2D DM with two nodes and $\Lambda = 1$ eV, which corresponds to graphene on the substrate (top panels), and ii) a hypothetical WSM with two nodes and $\Lambda = 1$ eV (bottom panels). We investigate selective ($g = 1$) and uniform ($g = 2$) pumping. In both cases, the values of $\alpha$ are chosen to be smaller than the equilibrium critical coupling: $\alpha < \alpha_c^{\text{CDW/RI}} \approx 1$ for graphene and $\alpha < \alpha_c^{\text{CDW/RI}}$ for WSM. According to Figures 2–5, this is the coupling regime in which pumping promotes the excitonic instability.

In Figure 6, the line separating the dark ($\Delta_{\text{max}} = 0$) and bright ($\Delta_{\text{max}} \neq 0$) regions of the phase diagram defines the dependence of $T_c$ on $\bar{\mu}$. The behavior of $T_c$ is similar in all cases considered: $T_c$ increases monotonically with the chemical potential until it reaches a maximum at a certain value of $\bar{\mu}$; further increasing $\bar{\mu}$ leads to a decrease in $T_c$ due to screening. The peak in $T_c$, as a function of $\bar{\mu}$ determines the regime in which optical pumping is most efficient. The value of the chemical potential that gives the largest $T_c$ is determined by the material specific constants $\alpha$ and $g$. For example, for a 2D DM and the same value of $\alpha$, the peak shifts to smaller values of $\bar{\mu}$ due to larger screening. For a WSM, the downturn in $T_c$ for $g = 2$ occurs at a slightly larger $\bar{\mu}$ compared to $g = 1$ due to different values of $\alpha$ considered in the two cases.

4. Discussion and Experimental Feasibility

4.1. Comparison to Equilibrium DMs with Population Inversion

Before discussing experimental feasibility of excitonic condensate in pumped DM, we should note that finite density of electrons and holes can be realized in equilibrium systems. In the 2D case, parallel magnetic field applied to a single layer of graphene creates two perfectly nested electron and hole Fermi surfaces, allowing for condensation. Finite equilibrium populations can be achieved in double layer systems, such as double layer graphene and TI thin films, where the chemical potentials of the two “layers” are tuned independently by using electrostatic gates.

A similar situation can be realized in materials with multiple Dirac/Weyl nodes, where the energies of at least two nodes are not required to be the same. A specific example in 2D is a topological Kondo insulator SmB$_6$ at finite chemical potential, provided there is a sufficient energy offset between the nodes at $\Gamma$ and $X/Y$ points. In 3D, perfectly nested electron and hole Fermi surfaces can be achieved in WSM with broken spatial inversion symmetry.

An important difference from earlier work on double layer 2D systems is that our theory of pumped DM applies to a single layer DMs, that is, monolayer graphene or TI surface. In this case, finite populations of electrons and holes cannot be made by gating alone. Furthermore, as will be shown below, the effect of metallic screening can be minimized in single layer DMs. In double layer graphene, the number of fermions involved with screening is twice that of single layer graphene and therefore the maximum value of the coupling constant decreases by a factor of 2, this sets an upper limit on the critical temperature of $1 \text{mK}$. For single layer DMs the maximum value of the coupling constant increases by at least a factor of 2 (4, in the absence of the valley degree of freedom, as in 3D TIs). This leads to an exponentially enhanced upper critical temperature relative to the double layer case. Furthermore, since the system is a single layer, the interaction is not exponentially suppressed with increasing the distance between layers as in the case of the double layer system.

More generally, optical pumping is a novel way of generating excitonic states that have a transient nature. In the case of
Figure 6. The maximum value of the excitonic gap as a function of $\bar{\mu}$ and $T$. Top row is for a 2D DM with $\alpha = 0.7$ for a) $g = 1$ and b) $g = 2$. Bottom row is for a 3D DM (WSM) for c) $g = 1$ and $\alpha = 2.5$ and d) $g = 2$ and $\alpha = 1$. Note that for WSM with $g = 1$, only the EI phase is realized. For WSM with $g = 2$ both EI and CDW phases are present, however the CDW gap is the largest. Hence, $\alpha$ is chosen to be smaller than the corresponding equilibrium critical coupling in panels (c) and (d). The color shade represents the absolute value of the gap in meV's. The energy cutoff is $\Lambda = 1$ eV in both 2D and 3D cases. Note that here we retain physical units for energy and temperature to make a connection with estimates for $\Delta_{\text{max}}$ and $T_c$ in Table 2.

DMs, it also offers a way of enhancing and effectively controlling the strength of interaction between non-equilibrium electron and hole populations.

4.2. Estimates for Excitonic Gaps and $T_c$ in Pumped DMs

Numerical estimates of the critical temperature and the size of the excitonic for several examples of realistic and hypothetical 2D and 3D DMs are summarized in Table 2.

The material specific parameters of the model that control the size of the gap and $T_c$ were extracted from experimental or ab initio data. These parameters are i) the dimensionless coupling constant $\alpha$, which is determined by the dielectric constant $\varepsilon$ and the Fermi velocity $v$, ii) the energy range for the Dirac states exist, that is, the cutoff energy scale $\Lambda$ of the effective Dirac Hamiltonian; this energy scale limits the range of the non-equilibrium chemical potentials $\bar{\mu}$ that can be achieved by pumping, and iii) the degeneracy factor $g$. In addition, we list the expected lifetimes $\tau$ of the transient excitonic state inferred from experimental data for existing 2D DMs. The values of the non-equilibrium chemical potentials could be estimated based on the density of photoexcited carriers and the properties of the laser pump in the experiment. Such analysis is provided for graphene, for which such experimental data are readily available in the literature. In other cases, we take $\bar{\mu} = \Lambda/2$.

Below we provide a more detailed explanation for the choice of material parameters and discuss the results presented in...
carrier densities in typical experiments can be estimated based on the following relation \( h\omega_{\text{pump}} n_{\text{exc}} \approx \Phi A_0 \) where \( \Phi \) is the pump fluence, \( h\omega_{\text{pump}} \) is the pump energy, and \( A_0 \) is the absorption coefficient of graphene. Taking \( A_0 = 0.02, h\omega_{\text{pump}} = 1 \text{ eV} \) and assuming \( n_{\text{ex}} = n_{\text{ex}} \approx n_{\text{ex}} \) we find that a chemical potential \( \mu \approx 500 \text{ meV} \) corresponds to carrier density \( n_{\text{ex}} \approx 10^{13} \text{ cm}^{-2} \) (and a pump fluence \( \Phi \approx 100 \mu \text{J cm}^{-2} \)), which can be achieved in present experiments.\(^{128}\)

As already mentioned in Section 2.1, optical excitation in 3D TIs is different from graphene and involves bulk states, since the energy of the pump pulse exceeds the insulating bulk gap (the bandgap in Bi\(_2\)Se\(_3\) is \( \approx 300 \) meV while \( h\omega_{\text{pump}} \approx 1.5 \text{ eV} \)). As a result, electrons are pumped from the valence band into the conduction band and then scatter down to the low-energy Dirac surface states. Recent trARPES experiments showed that the lifetime of the photoexcited carriers in the upper Dirac cone increases as the energy approaches the node.\(^{51,63}\) In samples where the chemical potential is close to the node, the relaxation bottleneck due to the vanishing phase space at the node leads to long-lived population inversion.\(^{65}\) Based on the reported trARPES results, the non-equilibrium chemical potential in 3D TIs is of the order of 100 meV.\(^{51}\)

For valley-pumped suspended graphene (\( \alpha = 2.2, g = 2 \) and \( \mu = 500 \text{ meV} \)), we estimate \( \Delta_{\text{max}} \approx 10 \text{ meV} \) and the corresponding \( T_c \approx 70 \text{ K} \), which is within the resolution of typical trARPES experiments. Other experimental probes that may be sensitive to the presence of excitonic gaps, are time-resolved optical conductivity\(^{45}\) and photoluminescence measurements, which are expected to show a peak due to enhanced photoluminescence from recombined electron-hole pairs.\(^{18}\) The calculation of optical conductivity in pumped 3D DMs in the normal and excitonic insulator states has been carried out in ref. 16.

Due to the large bulk dielectric constant, \( \alpha \) is typically smaller in 3D TIs compared to graphene. For the minimum value \( \alpha = 0.1 \), the estimated gap is only a fraction of a meV, that is, it is below the resolution of typical trARPES experiments. However, for values of \( \alpha \) closer to those in graphene on the substrate (0.4 \( \leq \alpha \leq 1 \)), a gap of few meV and \( T_c \) of tens of K can be obtained. Larger coupling constants could be found in materials with smaller dielectric constant or smaller Fermi velocities. For instance, Dirac states with \( \nu \approx 10^4 \text{ m s}^{-1} \) and \( \alpha \approx 7 \), were found in quasi-2D organic conductors.\(^{129,130}\)

We should note that on the timescale of the population inversion in graphene (100 – 200 fs), the local electronic temperature for the electron and hole distributions is still very large, for example, it reaches few thousand K.\(^{46,68}\) which greatly exceeds the predicted \( T_c \) even in the case of suspended graphene. This problem can be overcome in materials with increased lifetime of the population inversion such as (Sb\(_{1-x}\),Bi\(_x\))\(_2\)Te\(_3\)\(^{63}\) where the carriers have more time to cool down. It could be beneficial to use pumping with mid-infrared photons in order to minimize the initial temperature of photoexcited carriers. Another possibility is to search for materials with desired properties that maximize \( T_c \). Critical temperatures of hundreds of K can be achieved in 3D TIs with a single Dirac node, a large bandgap, and a relatively large \( \alpha \) (see estimates for a hypothetical 2D DM with \( g = 1 \) and other parameters similar to graphene).

A recent experimental study reported the evidence of excitonic superfluid phase on the surface of Bi\(_2\),S\(_x\)Se\(_3\) 3D TI.\(^{131}\) By

### Table 2. Estimates of the critical temperature \( T_c \) and the maximum of the excitonic gap \( \Delta_{\text{max}} \) in pumped 2D and 3D DMs.

| Material          | \( \alpha \) | \( \Lambda \) [eV] | \( \nu \) [ps] | \( T_c \) [K] | \( \Delta_{\text{max}} \) [meV] |
|-------------------|-------------|-----------------|-------------|-------------|-----------------|
| Graphene (substrate) | 0.4 – 1.0 | 1.0             | 0.1         | 0.1 for \( g = 4 \) | 0.1 for \( g = 4 \) |
|                  |             |                 |             | 6 – 35 for \( g = 2 \) | 5 – 1 for \( g = 2 \) |
| Graphene (suspended) | 2.2        | 1.0             | 0.1         | 2 for \( g = 4 \) | 0.3 for \( g = 4 \) |
|                  |             |                 |             | 70 for \( g = 2 \) | 10 for \( g = 2 \) |
| 3DTI             | 0.1 – 1.0  | 0.1             | 1 – 10      | 0 – 30      | 0 – 3           |
| 2D DM with \( g = 1 \) | 0.4 – 2.2  | 1.0             | – 50 – 500  | 10 – 100    |                |
| Cd\(_3\)As\(_2\) DM | 0.1        | 1               | 6           | 0.1         | 0.03            |
| TaAs WSM         | 1          | 0.2             | 2           | 0.3         |                |
| 3D DM with \( g = 1 \) | 1 – 3      | 1               | 1 – 20      | 0.3 – 3     |                |
| 3D DM with \( g = 2 \) | 1 – 3      | 1               | 10 – 60     | 1 – 10      |                |
| 3D DM with \( g = 4 \) | 1 – 3      | 1               | 1 – 2       | 0.1 – 0.3   |                |

Material-specific parameters, that is, the dimensionless coupling constant \( \alpha \), the cutoff energy scale \( \Lambda \), and the lifetime of the transient excitonic state \( \nu \) were estimated based on existing literature. For graphene and 3D TIs, the average chemical potential \( \mu \) used in the numerical calculation of the gap was estimated based on experimental data. For all other cases, the values of \( \Delta_{\text{max}} \) and \( T_c \) were obtained by taking \( \alpha = \Lambda / 2 \). The last row in the section of the table corresponding to 2D DMs shows the estimates for a hypothetical 2D DM with \( g = 1 \) and with other parameters similar to graphene. For hypothetical 3D DMs (last three rows of the table) the cutoff energy scale \( \Lambda = 1 \text{ eV} \) is assumed.

Table 2 for 2D and 3D DMs. We also discuss features of the experimental setup that could affect the detection of the transient excitonic states.

#### 4.2.1. 2D DMs: Graphene and Topological Insulators

The effective dielectric constant for graphene is given by \( \varepsilon = (\varepsilon_{\text{sub}} + \varepsilon_{\text{vac}}) / 2 \), where \( \varepsilon_{\text{sub}} \) is the dielectric constant of the substrate (vacuum). Using the reported values of \( \varepsilon \) for graphene on the substrate, \( \varepsilon \in [2.15] \) and taking \( \nu = 1 \times 10^4 \text{ m s}^{-1} \), we get 0.1 \( \leq \alpha \leq 1 \). For two typical substrates, SiC and SiO\(_2\), \( \alpha \approx 0.4 \) and \( \alpha \approx 0.8 \), respectively. For free-standing graphene, \( \alpha \approx 2.2 \). We consider conventional (uniform) pumping with linearly polarized light (\( g = 4 \)) and valley-selective pumping (\( g = 2 \)). In order to enable valley-selective pumping, a finite energy gap in the Dirac spectrum is required to generate an orbital magnetic moment in the valleys. A finite gap can be induced in graphene by inversion-symmetry breaking, for example, due to the substrate.

We consider 3D TIs with a single Dirac cone (\( g = 1 \)), such as Bi\(_2\)Se\(_3\) and related materials. The dielectric constant in 3D TIs is taken to be \( \varepsilon = (\varepsilon_{\text{T1}} + \varepsilon_{\text{vac}}) / 2 \), where \( \varepsilon_{\text{T1}} \) is the dielectric constant of the bulk. The bulk dielectric constant is of the order of 100 (\( \varepsilon_{\text{T1}} = 113 \) in Bi\(_2\)Se\(_3\), and \( \varepsilon_{\text{T1}} = 75 – 290 \) in Bi\(_2\)Te\(_3\))\(^{124}\), leading to \( \varepsilon \approx 50 \). It is possible that in some samples, \( \varepsilon \) can be reduced to \( \varepsilon \approx 30 \) due to heavy doping.\(^{125}\) Combining the values of \( \varepsilon \) and the typical velocities of the Dirac states \( \nu = 2 \times 10^8 \text{ m s}^{-1} \)\(^{126}\), we obtain \( \alpha \) in the range \( \alpha \in [0.1 : 0.4] \). However, since smaller values of \( \varepsilon \) can be found, for example, in the so-called “slow” DMs\(^{127}\) and \( \varepsilon \) could be tuned by gating in TI thin films, we consider a larger range \( \alpha \in [0.1 : 1.0] \).

It can be shown that in the case of graphene, \( \mu_{\text{exc}} = h\sqrt{\frac{\nu}{\pi n_{\text{exc}}}} \), where \( n_{\text{exc}} \) is the density of photoexcited carriers. Furthermore,
using scanning photocurrent microscopy, it was shown that photoexcited electrons and holes form charge neutral bound states which propagate over distances of 1 mm at temperatures up to 40 K. There are indications that the observed effect could be explained by the theoretical model discussed in this work. For instance, in Bi$_2$Sb$_2$Se$_3$ samples the Fermi level is close to the Dirac point which is an important condition for the formation of excitonic state. Moreover, the the longest lifetime of the population inversion was reported in Sn-doped 3D TIs.[63,64] There are no signatures of the excitonic phase in pure Bi$_2$Sb$_2$Se$_3$ in which these conditions are not satisfied.[131] Although the existence of Dirac point which is an important condition for the formation of an excitonic gap, graphene,[59,60,62] suppressed Auger recombination can result in a sizable excitonic gap in these systems. Therefore, we assume selective pumping and choose the value of g that maximizes the size of the gap and $\tau_c$. Such an optimistic estimate corresponds to $g = 2$ for a WSM and $g = 4$ for a DSM in the CDW phase. It is possible to consider pumping on a single Dirac or Weyl node, $g = 2$ and $g = 1$, respectively; however, in this case only the EI phase is realized leading to substantially smaller gaps.

The numerical estimates for the excitonic gap and critical temperature in 3D DMs are summarized in the lower half of Table 2. For TaAs WSM, we predict $\Delta_{\text{max}} \approx 0.3$ meV and $T_c \approx 2K$. Due to a small $\alpha$ resulting from a nearly dielectric constant, comparable to that of a 3D TI, the estimated gap and $T_c$ for Cd$_3$As$_2$ DSM is one order of magnitude smaller. Despite the small size, the predicted excitonic gap in these two types of 3D DMs is nevertheless finite. In contrast to this, the equilibrium excitonic gap is zero, since the material-specific coupling $\alpha$ is both examples is smaller than the equilibrium critical coupling. In the case of Cd$_3$As$_2$, we also include the estimate of the lifetime of the transient excitonic state, $\tau_c \approx 6$ ps based on experimental results of ref. [61].

The last three rows in Table 2 correspond to hypothetical 3D DMs with improved parameters, for example, 1 $\leq \alpha \leq 3$ and the cutoff energy scale $\Lambda$ for the Dirac/Weyl states of the order of 1 eV. As before, we consider selective pumping and a few values of the degeneracy factor, $g = 1, 2,$ and $4$. The largest gap and $T_c$ comparable to the values for selectively pumped suspended graphene, are predicted for the CDW phase with $g = 2$.

### 4.2.2. 3D DMs: Dirac and Weyl Semimetals

Although many 3D DMs have been discovered recently, in Table 2 two main examples are considered, Cd$_3$As$_2$ DSM and TaAs family of WSMs, that is, TaAs, TaP, NbAs, and NbP. For these materials, extensive ARPES data and electronic structure calculations are available, allowing for determination of the material-specific coupling constant. Thus, for Cd$_3$As$_2$, $v_e \approx v_h \approx 1.3 \times 10^8$ m s$^{-1}$, $v_e \approx 3.3 \times 10^5$ m s$^{-1}$,[132] $\Lambda \approx 1$ eV,[132] and $\varepsilon = 36$.[133] For the TaAs family, $v \approx 2.5 \times 10^5$ m s$^{-1}$,[134] $\Lambda \approx 200$ meV,[31,134] and $\varepsilon = 10$.[135] Note that in the case of anisotropic Dirac dispersion, we use the average velocity for numerical estimates. Based on these values, we estimate $\alpha = 0.1$ and $\alpha = 1$ for Cd$_3$As$_2$ and TaAs, respectively.

Typically, the degeneracy factor in realistic 3D DMs is large. Since screening increases with $g$, uniform pumping cannot generate a sizable excitonic gap in these systems. Therefore, we assume selective pumping and choose the value of $g$ that maximizes the size of the gap and $\tau_c$. Such an optimistic estimate corresponds to $g = 2$ for a WSM and $g = 4$ for a DSM in the CDW phase. It is possible to consider pumping on a single Dirac or Weyl node, $g = 2$ and $g = 1$, respectively; however, in this case only the EI phase is realized leading to substantially smaller gaps.

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The last three rows in Table 2 correspond to hypothetical 3D DMs with improved parameters, for example, 1 $\leq \alpha \leq 3$ and the cutoff energy scale $\Lambda$ for the Dirac/Weyl states of the order of 1 eV. As before, we consider selective pumping and a few values of the degeneracy factor, $g = 1, 2,$ and $4$. The largest gap and $T_c$ comparable to the values for selectively pumped suspended graphene, are predicted for the CDW phase with $g = 2$.

### 4.3. Dynamics of the Order Parameter

For the numerical estimates of the gap and $T_c$ presented in Table 2, we assumed a quasiequilibrium state with an infinitely long lifetime. In realistic systems, the population inversion is sustained over a finite time range and decays towards equilibrium with a single Fermi–Dirac distribution. As a result, the excitonic gap has a finite lifetime.

The basic condition for observation of the transient excitonic states is $\tau_c << \tau$, where $\tau_c = \hbar/\Delta$ is the timescale for the formation of the excitonic condensate and $\tau$ is the lifetime of the population inversion. Assuming $\tau \approx 100 - 200$ fs,[48] and $\Delta \approx 10$ meV in graphene,[49] and $\tau \approx 10$ ps,[63] and $\Delta \approx 1$ meV in 3D TIs, this condition is satisfied and therefore excitonic gaps could in principle be observed. Apart from being difficult to observe in experiment due to finite energy resolution, smaller gaps would require larger lifetimes of the population inversion.

In the case of pumped graphene, which has been studied extensively, the global carrier dynamics is governed by two main processes, the initial thermalization due to carrier-carrier and carrier-phonon scattering which occurs on the time scale of $50 - 100$ fs and further phonon-induced carrier cooling which happens on picosecond timescale.[136] The population inversion, if realized, develops within the first (fs) stage of the dynamics. It has been shown theoretically that the most important mechanism for build-up of population inversion is the phonon-induced intraband scattering, which leads to scattering of the highly excited carriers down toward the Dirac node resulting in the filling of the states at low energies. This process is responsible for a carrier relaxation bottleneck close the Dirac node and leads to population inversion. The decay of the population inversion on a 100 – 200 fs timescale, as observed in the experiment,[46] is caused by Coulomb-induced Auger recombination.[136] Neglecting Auger recombination processes in theoretical simulations based semiconductor Bloch equations leads to a long-lived population inversion, inconsistent with reported experimental values.

Despite graphene being the best candidates in terms of the size of the gaps and $T_c$, the lifetime of population inversion in typical hole-doped graphene on the substrate is relatively short (100 – 200 fs). Longer lifetimes are expected in undoped graphene with chemical potential at the Dirac node, where recombination of electron–hole pairs is suppressed due to vanishing phase space at the node. Continuous pumping could be used to sustain the population inversion over sufficiently long timescales, effectively leading to a quasiequilibrium excitonic state. However, in this scheme high temperatures of the photoexcited carriers will be destructive to excitonic condensation.

A recent theoretical work suggested that population inversion could be also realized in 3D DMs.[111] It was shown that ultrarelativistic electronic dispersion of WSMs leads to strong suppression of Auger recombination due to phase space restrictions imposed by energy and momentum conservation. Considering that the overall dynamics in 3D DMs is qualitatively similar to that of graphene,[59,60,62] suppressed Auger recombination can result in a population inversion on the timescales that are several orders of magnitude larger than in graphene and are comparable or larger than in 3D TIs.

It is important to mention the experimental work on Cd$_3$As$_2$ DSM, which used optical-pump THz-probe spectroscopy to

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**References:**

[131] Note that in the case of anisotropic Dirac dispersion, we use the average velocity for numerical estimates. Based on these values, we estimate $\alpha = 0.1$ and $\alpha = 1$ for Cd$_3$As$_2$ and TaAs, respectively.

[132] $\Delta_{\text{max}} \approx 0.3$ meV and $T_c \approx 2K$. Due to a small $\alpha$ resulting from a nearly dielectric constant, comparable to that of a 3D TI, the estimated gap and $T_c$ for Cd$_3$As$_2$ DSM is one order of magnitude smaller. Despite the small size, the predicted excitonic gap in these two types of 3D DMs is nevertheless finite. In contrast to this, the equilibrium excitonic gap is zero, since the material-specific coupling $\alpha$ is both examples is smaller than the equilibrium critical coupling. In the case of Cd$_3$As$_2$, we also include the estimate of the lifetime of the transient excitonic state, $\tau_c \approx 6$ ps based on experimental results of ref. [61].

[133] The last three rows in Table 2 correspond to hypothetical 3D DMs with improved parameters, for example, 1 $\leq \alpha \leq 3$ and the cutoff energy scale $\Lambda$ for the Dirac/Weyl states of the order of 1 eV. As before, we consider selective pumping and a few values of the degeneracy factor, $g = 1, 2,$ and $4$. The largest gap and $T_c$ comparable to the values for selectively pumped suspended graphene, are predicted for the CDW phase with $g = 2$.
investigate carrier relaxation close to the Fermi level.\[^{[61]}\] It was found that enhanced density of electrons and holes described by separate Fermi-Dirac distributions, is established via fast carrier-carrier and carrier-phonon interactions within 1 ps after the excitation. The relaxation of the population inversion is then dominated by electron-hole recombination on the characteristic timescale of 6 ps. These theoretical and experimental findings suggest that pumped WSM and DSM could be promising candidates for transient excitonic states, provided that other material parameters are such that screening effects are minimal.

Finally, the relaxation of the order parameter from the established exciton condensate state toward equilibrium can be studied using a time-dependent approach.\[^{[135]}\] based on semiconductor Bloch equations.\[^{[112,136–139]}\] In this approach we introduce electron and hole populations, \(n^e_k(t)\) and \(n^h_k(t)\), and the so-called interband polarization, or anomalous correlator, \(\mathcal{f}_k = \langle \epsilon^e_{-k} \epsilon^h_{k} \rangle\), which is related to the order parameter \(\Delta_k \equiv \Delta(k)\) as \(\Delta_k = \sum_k \mathcal{V}_k \mathcal{f}_k\) [see the definition of the gap in Equation (18)]. The dynamics of the single-particle expectation value is governed by the following system of differential equations

\[
\begin{align*}
\frac{dn^e_k}{dt} &= i\Delta^*_{-k} - i\Delta_k + \frac{dn^h_k}{dt}\Big|_{\text{scat}} \\
\frac{dn^h_k}{dt} &= i\Delta^*_{-k} - i\Delta_k + \frac{dn^e_k}{dt}\Big|_{\text{scat}} \\
\frac{df_k}{dt} &= i(\epsilon^e_{k} + \epsilon^h_{k})f_k + i\Delta'(t - n_k - n_k') + \frac{df_k}{dt}\Big|_{\text{scat}}
\end{align*}
\]

where \(df/dt|_{\text{scat}}\) denotes the phenomenological scattering terms.

The scattering terms can be calculated by coupling the electron and hole subsystems to featureless fermionic or bosonic reservoirs and by integrating out the reservoir degrees of freedom.\[^{[105,140]}\] In this simple model, we take into account interband and intraband scattering, characterized by the relaxation times \(T_1\) and \(T'_1\), respectively. The interband scattering leads to the recombination of electron-hole pairs, which results in the decay of the population inversion. Intraband scattering leads to thermal equilibration of electrons/holes within the corresponding Fermi-Dirac distributions described by the instantaneous chemical potentials \(\mu_{e(h)}(t)\) at time \(t\). The interband and intraband scattering terms are given by, respectively

\[
\begin{align*}
\frac{dn^e_k}{dt}_{\text{scat}} &= -\frac{n^e_k(t) - n^e_k(\mu_{e(h)}(t) - n^e_k(t))}{T_1} \\
\frac{df_k}{dt}_{\text{scat}} &= -\frac{f_k(t)}{T_2}
\end{align*}
\]

Note that both carrier-carrier and carrier-phonon scattering can contribute to intraband and interband processes. The model in Equation (29) does not differentiate between different relaxation paths but takes into account the overall intra- and interband relaxation with separate relaxation rates. Microscopic dynamical equations have been derived in the case of graphene, using the second-order Born–Markov approximation.\[^{[136]}\]

The relaxation of the gap is controlled by the dephasing time \(T_1\). It can be shown that it is related to the interband and intraband scattering times as \(T_{1}^{-1} = T_{1}^{-1} + T_{1}^{-1}\).\[^{[136]}\] Figure 7 shows the time-evolution of the order parameter, electron occupation, and non-equilibrium chemical potential for a 2D DM with parameters close to those of graphene on the substrate.

As the initial state at \(t = 0\), we take the values of the gap and electron and hole occupations calculated using the self-consistent gap equation, assuming the quasiequilibrium excitonic state with \(\bar{\mu} = \mu_e = -\mu_h\). At later times, the excitonic gap and the non-equilibrium chemical potentials are calculated self-consistently at each time step, by using Equation (28) and the definition of the gap. Note that here we focus only on the relaxation of the transient excitonic state toward equilibrium. Thus, we do not model the scattering processes associated with optical excitation and with building up of the population inversion on the fs timescales, for example, the interplay between carrier-phonon intraband scattering and Auger recombination in the case of graphene. To be consistent with the results for graphene, we fix

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**Figure 7.** Time evolution of a) the order parameter, b) the electron occupation, and c) the non-equilibrium chemical potential obtained from numerical integration of Equation (28) with scattering terms defined in Equation (29), for a 2D DM with \(T_1 = 100\) fs and \(T'_1 = 10, 100, \) and \(1000\) fs. Here \(g = 2,\) \(\alpha = 0.7,\) \(T = 0\). The value of the non-equilibrium potential before the relaxation \((t = 0)\) is \(\bar{\mu} = 500\) meV. \(\Delta_k\) and \(n^e_k\) are calculated at \(k = k_F\), where \(k_F\) is the Fermi wavevector in equilibrium. The inset in panel (b) shows the distribution of \(n^e_k\) at \(t = 0\). The dynamics of electrons and holes are identical to each other.
the interband scattering time to \( T_1 = 100 \text{ fs} \) and consider a few values for \( T_1' \). The simulation time is limited to 300 fs.

Since all relaxation channels contribute to the dephasing of the order parameter, the lifetime of the transient excitonic state is determined by the shortest of the relaxation times, or the largest scattering rate. This is illustrated in Figure 7. For the regime \( T_1' > T_1 \) (\( T_1 = 100 \text{ fs} \) and \( T_1' = 1000 \text{ fs} \) curves), the decay time of the non-equilibrium chemical potential \( \bar{\mu}(t) \), which determines the population inversion, is the same and is controlled by \( T_1' \). The relaxation of the gap (Figure 7a) and the electron/hole populations (Figure 7b) is slightly different in the two cases but it is also mainly governed by \( T_1' \) and is of the order of 100 fs. However, for the case \( T_1' < T_1 \), the gap decays to zero within tens of fs.

As mentioned above, experimental results\cite{Makhlin1996} and microscopic modeling\cite{Kamiya1999} suggest that the Coulomb-induced interband interactions, in particular Auger recombination, have the largest scattering rate and is the main source of decay of the population inversion on the timescale of 100–200 fs. Therefore, the main result of the simple dynamical model is that at least for the case of graphene, this gives an estimate for the lifetime of the transient excitonic state. More detailed experimental investigations and microscopic modeling similar to that done for graphene\cite{Kamiya1999} are needed to determine the scattering mechanisms and their contributions to the relaxation of the order parameter in other materials.

5. Conclusions and Outlook

In summary, we have reviewed the recent proposal for transient excitonic condensate in pumped DMs in the context of a broader search for dynamically-induced quantum states of matter. We have compared optically pumped DMs with other promising platforms for excitonic condensation, both in equilibrium and in non-equilibrium. The unique feature of DMs is the strongly energy-dependent DOS resulting from Dirac dispersion, which results in a tunable enhancement of the energy of the Coulomb interaction compared to the values accessible in equilibrium. We have shown using examples of 2D and 3D DMs that there exist a range of material parameters, in which optical pumping is more advantageous for excitonic condensation compared to equilibrium.

We have discussed the properties of the transient excitonic condensate, for example, spectroscopic signatures and the complex phase diagrams that result from the competition between electronic screening effects and the enhancement of DOS by pumping. The key features that characterize these exotic states are energy gaps in the quasiparticle spectrum which open up at the non-equilibrium chemical potentials of photoexcited electrons and holes. Such gaps and the corresponding suppression of spectral weight near the gaps could be detected by photoemission spectroscopy. Numerical estimates for specific materials indicate that the largest effect (gaps of the order of 10 meV and \( T_1 \) up to 70 K) could be achieved in graphene. 3D TIs with a single Dirac cone, in particular \((\text{Sb}_{1-x}, \text{Bi}_x)\text{Te}_3\) and similar compounds, are also promising candidates due to prolonged lifetime of the transient population inversion.\cite{Amin2010}

Recently, transport signatures of the excitonic condensate phase have been found on 3D TI surfaces, with critical temperature consistent with the values predicted by the theoretical model.\cite{Neto2009} Although the existence of the condensate requires rigorous confirmation, this work gives an incentive to use 3D TIs as a model system for further investigation of the transient excitonic states. One interesting problem is the effect of spin-momentum locking on the spin texture of the excitonic states and their dynamics. Furthermore, it was pointed out in previous theoretical work that non-equilibrium excitonic condensate in pumped semiconductors should exhibit persistent, self-sustained oscillations in the induced photo current and in the dynamics of the order parameter.\cite{Berggren2017,Engelbrecht2018,Engelbrecht2018a} A fascinating question that can be addressed using the theoretical model of pumped DMs augmented by dynamical equations for the order parameter, is whether there is an analogue of such effect in pumped semimetals with Dirac dispersion.

Finally, we proposed general guidelines to search for promising material candidates, which include i) large coupling, which requires small Fermi velocity and small dielectric constant, ii) small Dirac cone degeneracy, which is crucial for reducing screening effects, and iii) large \( \Lambda \) (the energy extent of the Dirac states). In addition, long-lived population inversion should be possible to allow for observation of the excitonic states on experimental timescales. Based on these criteria, we considered several hypothetical 2D and 3D systems, in which large gaps and critical temperatures comparable to those found in graphene could be achieved. Given the high pace of material discovery facilitated by high-performance material informatics, it is likely that more candidates will be identified. Combined with continuing advances in time-resolved spectroscopies, we believe that the ongoing search for transient collective states in Dirac materials will remain an active topic of research.

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Conflict of Interest

The authors declare no conflict of interest.

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