Abstract

Conventional Rabi oscillations are studied using the rotating wave approximation (RWA). It is an approximation used for the approximate analytic solution of time-dependent Schrödinger eqn. of a two-level system coupled to a weak electric field in resonant with the transition. In this approximation, we remove rapidly oscillating terms of the effective Hamiltonian, but its validity is limited to only when the internal frequency of the two-level system is close to the external frequency of field, i.e. in case of resonance. Far from conventional resonance, only relativistic fermionic systems exhibit a new type of Rabi oscillation, which is absent in conventional semiconductors and two-level systems. Our aim is to study nonlinear optics of these systems at the Weyl (Dirac) node, since all interesting physics at low energy occurs in the vicinity of the Weyl(Dirac) node - particularly the phenomenon of anomalous Rabi oscillation (ARO), which has been predicted theoretically in single layer graphene by our group (and also by others where these ideas go under the name of "Floquet theory"). In order to study this, we employ an approximation known as asymptotic rotating wave approximation (ARWA). We have studied this phenomenon as well as its conventional counterpart that occurs close to resonance (ordinary Rabi oscillations) and verified the claims numerically.

Graphene is a prototype of a two-dimensional massless Dirac-fermion system. The three-dimensional (3D) analog of graphene is called a Weyl semimetal. 3D topological insulators (TI) have surface states that consist of massless 2D Dirac fermions. These three systems have some features in common - in addition to being composed of massless fermions, they possess spin-like internal degrees of freedom. Such relativistic fermion systems are best characterized using the phenomenon of anomalous Rabi oscillation (ARO) that occurs far from conventional resonance and is unique to these systems. These oscillations are absent in two-level atoms and conventional semiconductors. The main difference in the non-linear optical response between graphene-like materials and Weyl semimetal (or surface state states of TI) is the latter's pronounced anisotropy in the wave vector dependence of both the conventional and anomalous Rabi frequencies. A detailed analysis of this anisotropy shows that in Weyl metals (or on TI surface), the Rabi frequency has a form that resembles massless Dirac particles at certain points in the reciprocal space. Hence, in these systems, both the quasiparticles and the collective modes that exist in them in response to time-varying fields are ultra-relativistic in nature whereas in graphene only the original quasiparticles are massless, but the Rabi modes are massive. On surface states of TI, this anisotropy may be attributed to

the Zeeman term. It is shown that the Zeeman term has no qualitative effect on the conventional Rabi frequency, but its absence makes the anomalous Rabi modes massless. A fully numerical solution of the Floquet-Bloch equations unequivocally establishes the presence of not only anomalous Rabi oscillations in these systems but also their massless character.

In two-level systems, it is recognized that conventional Rabi oscillations undergo a shift in the resonant frequency which is the Bloch-Siegert shift. This shift is quite small in such systems and hence largely of academic interest. The present work shows that in graphene, such shifts can be substantial - especially upon the inclusion of band curvature effects and Rashbha coupling. Both conventional and anomalous Rabi oscillations are resonance phenomena, in other words, the amplitude of these oscillations peak when the condition for resonance is obeyed. The purpose of study the shift in the conditions for resonance brought about by **(a)** the presence of strong fields upon the inclusion of frequency doubling effects or **(b)** band anisotropy due to next nearest neighbor hopping or Rashba effect. We refer to both these shifts as Bloch-Siegert shifts, although only **(a)** was envisaged by Bloch and Siegert.

In science, verification of a theory is usually accomplished by performing a suitable experiment. We have described pump-probe spectroscopy, which is used to experimentally probe the phenomenon of anomalous Rabi oscillations - a central idea in this thesis. In this experiment, two successive laser pulses are used, one for exciting the system in a certain way, called pump pulse and other to test it after a variable time delay, called probe pulse. This simply involves looking for periodic oscillations in the differential transmission coefficient versus pump pulse duration (alternatively, the area of the pump pulse). When all else remains fixed (including pump-probe delay), these plots exhibits oscillations with a frequency corresponding to the anomalous Rabi frequency. Furthermore, the amplitude of these oscillations decay as a power law in the pump duration with a characteristic exponent that is indicative of the particular system under consideration.

Furthermore, it is shown that ARO exists even if the EM field is treated quantum mechanically. This establishes that the AROs are not due to approximations or assumptions we have made. By using probability amplitude equation, we have studied anomalous Rabi oscillations in the presence of a quantum field. Finally, we described limiting case, how these quantum result will convert in the classical result. New phenomena such as zero-point or vacuum anomalous Rabi oscillations are seen. Therefore, it is no exaggeration to say that the anomalous Rabi oscillation is a phenomenon that is uniquely suited to study condensed matter realizations of relativistic systems.