

COHERENT CONTROL OF ELECTRONIC STATES AND QUANTUM DYNAMICS OF MATTER BY EXTERNAL FIELDS

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 We present a theory of optical absorption for the renormalized band structure of semiconductors under irradiation by an intense off-resonant infrared laser. It is shown that there appears an induced absorption band below the original band gap, in agreement with a recent experiment. We also present some results on the coherent control of quantum dynamics using external time-dependent fields.

1. INTRODUCTION

The coherent manipulation of electron wave functions and quantum dynamics of matter such as molecules, solids or nano structures is one of the central issues not only in the physics but also in the field of advanced technology. One of the motivation is, of course, the potential application to the future devices based on the principle of quantum mechanics. For example, the development of techniques to manipulate assemblies of two-level systems (qubits) is a crucial requirement in the architecture of quantum computers. The attempt to control chemical reactions by using intense electromagnetic fields is another topic in this connection. Theoretically, the study of quantum states and dynamics of externally driven systems poses a new class of problems. In the present paper, we will review recent progress in the study of coherent control of electronic states of matter and their dynamics both from theoretical and experimental side.

2. BAND GAP RENORMALIZATION OF SEMICONDUCTORS BY INFRARED LASER

Recently, it was reported that the apparent band gap of GaAs crystal is reduced to a remarkable extent under irradiation of intense off-resonant infrared laser[1]. This is

 $H = H_0 + H_I(t),$

a coherent phenomenon in the sense that it does not accompany with any real excitation of carriers. It is a strongly nonlinear optical process, well beyond a perturbation theory. From application side, this finding means a possibility to construct efficient nonlinear optical devices which have a very short response time.

This effect is related with the so-called dynamic localization of carriers under oscillating electric field[2]. However, it should be noted that the dynamical localization only results in the shrinking of band width, and the apparent band gap would expand in contradiction with experimental observation. In order to explain the observed data, we must take into account the fact that the band gap of semiconductors is determined by the intratomic and interatomic exchange (transfer energy). In other words, we must devise a theoretical framework of band calculation that takes account of the influence of the laser field from the beginning.

Here, we present a theory of laser-induced coherent band renormalization, and show a result of calculated optical absorption spectrum. First we show results for a one-dimensional model. The one-dimensional model is best suited to see the essential features of the problem.

The Hamiltonian is given by

$$
- (1)
$$

$$
H_0 = -t_1 \sum_n (n, 2\lambda n, 1| + |n, 1\lambda n, 2|) - t_2 \sum_n (n, 2\lambda n + 1, 1| + |n + 1, 1\lambda n, 2|)
$$
 (2)

$$
H_I(t) = -eE(t)\left\{\frac{\alpha}{2}\sum_n \left(n, 2\lambda\left(n, 2\right) - \left|n, 1\lambda\left(n, 1\right)\right| + \beta \sum_n n\left(n, 1\lambda\left(n, 1\right) + \left|n, 2\lambda\left(n, 2\right)\right|\right)\right\},\tag{3}
$$

where $|n,1\rangle$ and $|n,2\rangle$ are s-p hybridized orbitals at n-site, which are defined as linear combinations of the s and p orbitals at n-site, $|n,s\rangle$ and $|n,p\rangle$, as

$$
|n,1\rangle = (n,s\rangle - |n,p\rangle)/\sqrt{2},\qquad(4)
$$

$$
|n,2\rangle = (n,s\rangle + |n,p\rangle) \sqrt{2}
$$
 (5)

The intraatomic transfer energy t_1 is then given by $t_1 = (\varepsilon_p - \varepsilon_s)/2$ with the energies of the s and p orbitals, ε_s and ε_p , and t_2 is the interatomic transfer energy. $E(t)$ is the electric field of the infrared laser which is oscillating sinusoidally, and α and β are the parameters for the intraatomic and interatomic Stark effect. In accordance with the dynamical Stark effect, the energy levels of each atom oscillate up and down as shown in Fig. 1.

The eigenvalues of the unperturbed Hamiltonian H_0 are given by

$$
E_k^{\pm} = \pm \sqrt{t_1^2 + t_2^2 + 2t_1t_2 \cos ak} \,, \tag{6}
$$

where *a* is the lattice constant and *k* is the wave vector. There appear a conduction band and a valence band with the band gap energy given by $2 | t_2 - t_1 |$. The interaction Hamiltonian is written as

 $H_1(t) = -eE(t)(\alpha x/2 + \beta x)$ (7)

where *x* and *X* are the intraatomic and interatomic displacement operators given by

$$
x = \sum_{n} (n, 2\lambda n, 2| - |n, 1\lambda n, 1|)
$$

$$
X = \sum_{n} n(n, 1\lambda n, 1| + |n, 2\lambda n, 2|)
$$
 (8)

respectively. Note that this Hamiltonian violates the translational symmetry. It can be recovered by applying the time-dependent gauge transformation with the unitary operator,

$$
U(t) = \exp\{-ieA(t)(\alpha x/2 + \beta X)\}\tag{9}
$$

where $A(t)$ is a gauge potential,

$$
A(t) \equiv \int_{-L}^{t} E(\tau) d\tau. \tag{10}
$$

The optical absorption spectrum around the band gap energy is measured by the near infrared or visible light. The interaction with this probe light can be treated by the lowest order perturbation theory. However, we cannot rely upon Fermi's golden rule to calculate the transition probability, because the system is under time dependent strong perturbation and is far from the thermal equilibrium. In order to overcome this difficulty, we have developed a time-dependent generating function formalism. The time-averaged absorption spectrum *I*(Ω) for the probe photon with energy Ω is then given in the form

$$
I(\Omega) = \lim_{T_P \to \infty} \frac{1}{T_P} \int_0^{T_P} \int_{-\infty}^{+\infty} dt G(T, t) e^{-i\Omega t}
$$
 (11)

where $G(T, t)$ is a double-time Green's function (generating function). In order to calculate $G(T, t)$ actually, Bloch's theorem in the space domain coupled with Floquet's theorem in the time domain works efficiently.

In Fig.2, the calculated absorption spectrum is shown. The thin dotted line corresponds to the interband absorption for the unperturbed system. The bold dotted line represents the absorption spectrum under the irradiation of the infrared laser. The absorption spectrum is decomposed into contributions from the transitions with the change of Floquet indices n as shown in Fig.2. As can be seen from the figure, the intense laser field gives rise to two effects, namely, the shrinkage of the conduction band and the emergence of a new absorption band below the band edge. The latter originates from the transition between the Floquet states with different indices. In other words, it comes from the interband transition accompanying the simultaneous absorption of the photons of the infrared laser. This additional structure corresponds to the apparent shrinkage of the band gap. Note that this is a bulk effect so that the transmission of light below the edge region is strongly reduced as observed in the experiment[1].

The model has been extended to the three-dimensional system that corresponds to the electronic states of realistic semiconductors such as Si and GaAs. The valence band and the conduction band have been constructed by a tight-binding model with basis functions 2s and 2p. The Bloch states are described by the linear combinations of the hybridized orbitals as usual. The effect of the intense infrared laser is taken into account as an oscillation electric field as in the one-dimensional model. The gauge transformation is again applied for three-dimensional system that recovers the apparent translational symmetry of the crystal. The interaction with the probe light is treated by the lowest order of perturbation.

In Fig.3, the unpertubed band structure of GaAs is shown. The parameter values are chosen from literatures[3]. It is well known that the tight binding calculation generally gives the band gap energy larger than that of real materials, if one adopts the raw values of atomic data. Here we show the calculated results without any renormalization. In Fig.4, we show a result of the induced transmission change at the band gap calculated for parameters corresponding to GaAs. In the inset, the experimental data[1] for the differential transmission for GaAs is shown.

As shown here, there appears negative gain (i.e. induced absorption) below the band gap energy in the presence of intense infrared laser. Likewise, there occurs an increase of transmission above the band gap. The calculated line shapes of the differential transmission well reproduce the experimental features. The effect of the infrared laser has thus two distinct effects: the one is the reduction of the band width and the other is the appearance of induced absorption below the band gap. We may call the electronic state that is strongly mixed with photons (oscillating fields) a dressed semiconductor.

Fig. 4

3. COHERENT CONTROL OF QUANTUM DYNAMICS

As noted above, the band gap renormalization is closely related with the so called dynamic localization of electrons. This is the phenomenon predicted theoretically by Dunlap and Kenkre[2], who pointed out that the quantum migration of an electron in a one-dimensional lattice may be suppressed completely by applying oscillatory electric field with a suitable ratio of the frequency and amplitude. On the other hand, it has been shown by Grossmann et al.[4] that the quantum tunneling in a two-level system is suppressed in the oscillatory field with a suitable parameter values. This is called the coherent destruction of tunneling (CDT). One of the present author[5] pointed out that the CDT can be regarded as a special case of destructive interference between the transition paths of a two-level system, which can be understood clearly by use of the Landau-Zener theory [6,7] of nonadiabatic transitions. The dynamic localization and the CDT may be classified into a class of time-dependent problems that can be solved exactly or quasi exactly.

The recent success in the observation of a coherent oscillation of an electron in double quantum dots (QDs) of semiconductors[8] enhanced the motivation to utilize the QDs as elements of new information processing

devices (i.e. qubits) based on the principle of quantum mechanics. Similar phenomena have been reported earlier for Josephson qubits[9]. The establishment of the techniques of coherent manipulation of electrons is an essential requirement for the development of such quantum devices.

So far, the experimental realization of coherent manipulation of the electrons utilized the Rabi oscillation induced by the sudden changes of the external fields. For example, in order to transfer an electron fron the left dot to the right dot, one applies a rectangular gate pulse that switches the two level system between the resonant and off resonant condition suddenly. However, this requires a very high frequency components in the gate voltage fields, which may pose some difficulty in the future application.

Recently Saito and one of the authors[10] proposed a novel method of coherent manipulation that utilizes the quantum interference in the nonadiabatic transitions. According to this technique, one needs only smoothly varying gate pulses without sudden change. Let us consider electronic states in a simple double QDs, the Hamiltonian of which is given by

$$
H(t) = \varepsilon_1(t)|1\rangle\langle 1| + \varepsilon_2|2\rangle\langle 2| + \gamma(|1\rangle\langle 2| + |2\rangle\langle 1|)
$$
 (12)

where $|1\rangle$ and $|2\rangle$ describe the state in which the electron occupies a discrete level of the left and the right dot, respectively. The tunneling matrix element is given by γ . Without loss of generality we can assume that only the energy level of the state $|1\rangle$ changes. Let us assume that the energy level change as shown in Fig.5, namely the diabatic state $|1\rangle$ crosses $|2\rangle$ twice from $t = 0$ to $t = t_f$. If the magnitude of the tunneling parameter is amall enough as is usually the case, the transition is localized around the two level-crossings. In such a situation, the analysis by the tranfer matrix [5] is useful. The whole temporal evolution can be decomposed into a succession of the nonadiabatic transitions at the crossings and the free propagation in between. The transition probability q at each crossing is given by the cerebrated Landau-Zener formula [6,7].

In order to attain the maximum controllability, we adjust the speed of the energy change at crossing to give $q=1/2$.

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Then it can be shown that the transfer matrix for the two level system after the double crossing is given by

$$
K = \begin{pmatrix} e^{-i\phi} \cos \frac{\Theta}{2} & -i \sin \frac{\Theta}{2} \\ -i \sin \frac{\Theta}{2} & e^{i\phi} \cos \frac{\Theta}{2} \end{pmatrix}
$$
(13)

where $\Theta = S + 2\phi$ in which *S* is the dynamical phase proportional to the erea shown in Fig.5, and ϕ is the Stokes phase. The above formula means that we can obtain arbitrary unitary gate simply by adjusting *S* .

In Fig.6, the temporal profile of the existence probability of the electron in the left QD (solid line) and the right QD (dashed line) is shown for the value $S + 2\phi = 3\pi$, which corresponds to the complete transfer. As shown here, the agreement of the analytical formula and the numerical result is perfect. In the same way, we can manipulate an electron and transfer it from QD to QD for an array of QDs.

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