Dynamical collapse of the Peierls gap by an intense laser field

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The coherent interaction of electronic systems of matter with an intense radiation field strongly modifies the level scheme. In this work, it is shown that the application of an intense and high-frequency laser field on quasi-one-dimensional systems with Peierls gap will transiently reduce the gap. This is due to the dynamical reduction in the Bragg reflection that is responsible for the gap formation. The condition for the complete collapse of the Peierls gap is given as a function of the intensity and the energy quantum of the radiation, and the bandwidth of the original metallic phase of the quasi-one-dimensional system.

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When the intensity of a laser field becomes extremely high, the picture of individual photons is no longer adequate to describe the interaction with matter: we should rather go back to the classical picture of an oscillating electromagnetic field.[1] In such a case, the electronic states and the radiation field are strongly mixed each other to form *dressed states*.[2] In this Brief Report, we study the effect of intense laser field on the electronic states of solids. Specifically, we propose a possibility of coherent control of band gaps of Peierls insulators by a high-intensity, high-frequency electromagnetic field. It is predicted that the band gap will generally be reduced by the coherent interaction with the oscillating field and even be collapsed transiently if the parameter values of the radiation field are appropriately tuned.

The effect of high-density excitation of carriers in Peierls insulators is a subject of extensive studies in recent years. It has been shown that real excitation of carriers by femtosecond laser pulses induces various ultrafast phase change dynamics in crystals.[3,4] Our purpose here is, in contrast to these works, to propose a new mechanism of *coherent* modulation of the electronic states of matter by a high-intensity laser field. This work is a counterpart of our previous one,[5] in which it was shown that the apparent band gap of covalent semiconductors *expands* under an intense *low-frequency* electromagnetic field, as actually observed in GaAs irradiated by an infrared laser beam.[6]

In quasi-one-dimensional systems of solids, the metallic phase is generally unstable against the lattice distortion with wave vector $2k_F$, where k_F is the Fermi wave vector. The lattice will spontaneously deform and stabilize the total system by making a gap (Peierls gap) at the original Fermi edge.[7] This type of gap formation is widely recognized in quasi-one-dimensional systems, from TaS_3,[8] polyacetylene[9] to tetrathiafulvalene-tetracyanoquinodimethane (TTF-TCNQ),[10] to name but a few. The mechanism of the reduction of the gap presented below is rather general and is independent of the details of the interaction. For the sake of definiteness, we adopt here a model of bond-alternation-type insulators described by Su-Schrieffer-Heeger's (SSH) Hamiltonian.[9] We consider a linear chain of N atoms with a single atomic orbital on each of them. The Coulombic interaction between the valence electrons is neglected. Thus we adopt a spinless fermion model with half-filling.

Following SSH, it is assumed that the nearest-neighbor hopping integrals of the chain depend on the distance between the neighboring atoms. The Hamiltonian is given by

$$H = H_0 + V + H_L, \tag{1}$$

$$H_0 = -\frac{B}{2}\sum_j (|j+1\rangle\langle j| + |j\rangle\langle j+1|), \tag{2}$$

$$V = \frac{\alpha}{2}\sum_j (Q_{j+1} - Q_j)(|j+1\rangle\langle j| + |j\rangle\langle j+1|), \tag{3}$$

$$H_L = \sum_j \frac{Q_j^2}{2} \tag{4}$$

in which $|j\rangle$ is the Wannier function localized at jth atom, B is the half bandwidth in the hypothetical metallic state, Q_j represents the uniaxial displacement of the jth atom as shown in Fig. 1, and α is the electron-lattice coupling constant. The summation of j runs over N sites with an appropriate boundary condition and the number of spinless electrons is $N/2$. Actually, we consider the limit $N \to \infty$.

By the usual transformation to the Bloch states, $|k\rangle = N^{-1/2}\Sigma_j |j\rangle e^{ikaj}$ with a being the lattice constant in the metallic phase, H_0 is diagonalized as $H_0 = \Sigma_k \epsilon(k)|k\rangle\langle k|$, with $\epsilon(k) = -B\cos ka, (-\pi/a \le k \le \pi/a)$. The Bloch states are occupied for $-k_F \le k \le k_F$ with $k_F = \pi/2a$. At low temperatures, the metallic phase is unstable against the lattice deformation given by $Q_j = (-1)^j Q$. For this pattern, the interaction V is rewritten as

$$V = \alpha Q \sum_k \left(\left| k + \frac{\pi}{a} \right\rangle \langle k| e^{-iak} + |k\rangle \left\langle k + \frac{\pi}{a} \right| e^{iak} \right). \tag{5}$$

This form clearly indicates the origin of the band gap. For a fixed value of Q, V induces the Bragg reflection of the conduction electron, which mixes the state $|k\rangle$ and $|k+\frac{\pi}{a}\rangle$. The eigenvalues of the total Hamiltonian is obtained by solving the secular equation

$$\begin{vmatrix} \epsilon(k) - E & 2\alpha Q e^{iak} \\ 2\alpha Q e^{-iak} & \epsilon(k+\pi/a) - E \end{vmatrix} = 0 \tag{6}$$

from which the gap at $k = \pm k_F$ is obtained as $2|\alpha Q|$.

The total electronic energy $E_t(Q)$ per a unit cell is given by $E_t(Q) = \int_0^{k_F} E^{(-)}(k,Q)dk$, where $E^{(-)}(k,Q)$ is the lower

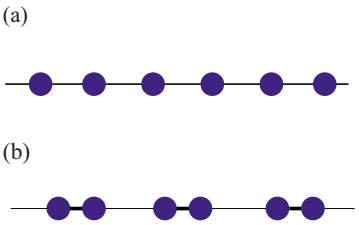

FIG. 1. (Color online) Bond-alternation model of the Peierls insulator for (a) metallic phase and (b) insulator phase.

branch of the solution of Eq. (6). The equilibrium value of the deformation Q_0 is then given by the solution of the equation

$$\frac{\partial E_t(Q)}{\partial Q} + \frac{\partial}{\partial Q}\frac{Q^2}{2} = 0. \tag{7}$$

The above equation may be solved approximately, by assuming the linear dispersion relation for $\epsilon(k)$ at $k \simeq k_F$. This enables us to obtain Q_0 and the energy gap $E_g^{(0)}$ in the ground state as a function of B and α.

Next, we introduce the interaction with the intense monochromatic laser field. Although the dynamical responses of the total electron-lattice system should be formulated, we consider here, as a first step, mainly the stationary response of the electronic system to the oscillating field for a fixed value of Q at the ground state Q_0. The electric field is assumed to be parallel with the chain direction. In the semiclassical picture with the long-wavelength approximation, the interaction Hamiltonian is written as

$$H_I(t) = eaE_0 \cos\,\omega t \sum_j j|j\rangle\langle j|, \tag{8}$$

where $e(>0)$ is the magnitude of the electronic charge and E_0 is the amplitude of the electric field oscillating with the frequency ω. It is assumed that the amount of the lattice displacement is small as compared with a. It is convenient to treat $H_0+H_I(t)$ as the unperturbed Hamiltonian and V as a perturbation. The point is that the Schrödinger equation driven by $H_0+H_I(t)$ can be solved exactly.[11,12] Specifically, the unitary operator for the time evolution is explicitly given by[13]

$$\begin{aligned} U(t) &= \exp_+\left\{-\frac{i}{\hbar}\int_0^t [H_0 + H_I(\tau)]d\tau\right\} \\ &= \exp\left[-i\frac{eE_0a}{\hbar\omega}\sin\,\omega t \sum_j j|j\rangle\langle j|\right] \\ &\times \exp\left[-\frac{i}{\hbar}\sum_k \int_0^t \epsilon_k(\tau)d\tau |k\rangle\langle k|\right], \end{aligned} \tag{9}$$

where $\epsilon_k(\tau) = -B\cos(ka - \frac{eE_0a}{\hbar\omega}\sin\,\omega\tau)$.

The Schrödinger equation for the state vector $|\Psi(t)\rangle$

$$i\hbar\frac{d}{dt}|\Psi(t)\rangle = \{H_0 + H_I(t) + V\}|\Psi(t)\rangle$$

is then transformed into the form

$$i\hbar\frac{d}{dt}|\psi(t)\rangle = \tilde{V}(t)|\psi(t)\rangle,$$

where $|\psi(t)\rangle = U^\dagger(t)|\Psi(t)\rangle$ and $\tilde{V}(t) = U^\dagger(t)VU(t)$. From Eqs. (5) and (9), $\tilde{V}(t)$ is given by

$$\begin{aligned} \tilde{V}(t) = \alpha Q_0 \sum_k \Bigg\{ &\exp\left[-2\frac{i}{\hbar}\int_0^t \epsilon_k(\tau)d\tau - \frac{ieE_0a}{\hbar\omega}\sin\,\omega t\right] \\ &\times \left|k + \frac{\pi}{a}\right\rangle\langle k|e^{-iak} + \text{H.c.}\Bigg\}. \end{aligned} \tag{10}$$

Expanding $|\psi(t)\rangle$ as $|\psi(t)\rangle = \Sigma_k C_k|k\rangle$, we obtain simultaneous equations for $C_{\pi/2a}(t)$ and $C_{-\pi/2a}(t)$

$$\begin{aligned} i\hbar\frac{d}{dt}C_{\pi/2a} &= \left\langle \frac{\pi}{2a}\middle|\tilde{V}(t)\middle|-\frac{\pi}{2a}\right\rangle C_{-\pi/2a}, \\ i\hbar\frac{d}{dt}C_{-\pi/2a} &= \left\langle -\frac{\pi}{2a}\middle|\tilde{V}(t)\middle|\frac{\pi}{2a}\right\rangle C_{\pi/2a} \end{aligned} \tag{11}$$

with

$$\begin{aligned} &\left\langle \frac{\pi}{2a}\middle|\tilde{V}(t)\middle|-\frac{\pi}{2a}\right\rangle \\ &= 2i\alpha Q_0 \exp\left[-\frac{2iB}{\hbar}\int_0^t \sin\left(\frac{eE_0a}{\hbar\omega}\sin\,\omega\tau\right)d\tau \right. \\ &\left. - \frac{ieE_0a}{\hbar\omega}\sin\,\omega t\right]. \end{aligned} \tag{12}$$

There are four characteristic scales of energy; the half bandwidth B, the Stark energy difference between the nearest-neighboring atoms eE_0a, the energy quantum of the oscillating field $\hbar\omega$, and the Peierls gap energy $E_g^{(0)}$. We assume, in the present work, that the inequality $B \gg \hbar\omega \gg E_g^{(0)}$ is satisfied. Furthermore, it is assumed that $\hbar\omega > eE_0a$. Then, Eq. (12) is approximated as

$$\begin{aligned} \left\langle \frac{\pi}{2a}\middle|\tilde{V}(t)\middle|-\frac{\pi}{2a}\right\rangle &\approx 2i\alpha Q_0 \exp\left[-\frac{2iB}{\hbar}\frac{eE_0a}{\hbar\omega}\int_0^t \sin\,\omega\tau d\tau\right] \\ &= 2i\alpha Q_0 \exp\left[-\frac{2iB}{\hbar\omega}\frac{eE_0a}{\hbar\omega}\right] \\ &\times \sum_{l=-\infty}^{\infty} J_l\left(\frac{2B}{\hbar\omega}\frac{eE_0a}{\hbar\omega}\right)e^{-il\omega(t+\pi/\omega)} \end{aligned} \tag{13}$$

in which $J_l(x)$ is the Bessel function of order l. We observe here that, in Eq. (11), the rapidly oscillating factors have been separated out in the phase factor. Since $\hbar\omega \gg E_g^{(0)}$ $(=2|\alpha Q_0|)$, the change in the variables $C_{\pm\pi/2a}(t)$ during a cycle of oscillation of the field is negligible. So we can safely approximate the coefficients in the right-hand side by an average over a period of oscillation

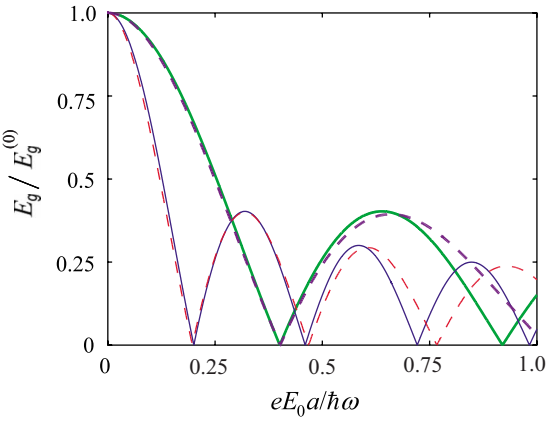

FIG. 2. (Color online) Reduction in the band gap $E_g/E_g^{(0)}$ of the Peierls insulator plotted against $eE_0a/\hbar\omega$ for two values of the energy quantum of the oscillating field, $\hbar\omega/E_g^{(0)}=2.5$ (thin lines) and $\hbar\omega/E_g^{(0)}=5.0$ (bold lines). The half bandwidth of the metallic phase is $B/E_g^{(0)}=15.0$. The analytical values of the formula (18) are plotted by the solid lines and the numerical results by the dashed lines.

$$\frac{\omega}{2\pi}\int_0^{2\pi/\omega}\left\langle \frac{\pi}{2a}\left|\tilde{V}(t)\right|-\frac{\pi}{2a}\right\rangle dt = 2i\alpha Q_0 \exp\left[-\frac{2iB}{\hbar\omega}\frac{eE_0a}{\hbar\omega}\right]J_0\left(\frac{2B}{\hbar\omega}\frac{eE_0a}{\hbar\omega}\right).$$

This is an inverse adiabatic approximation, which has been used to derive the formula for the coherent destruction of tunneling (CDT) (Ref. 14) in the two-level system.[15] We find that the effective gap is given by

$$E_g = \left|J_0\left(\frac{2B}{\hbar\omega}\frac{eE_0a}{\hbar\omega}\right)\right| E_g^{(0)}. \tag{14}$$

The band gap is thus reduced by the factor $\left|J_0(\frac{2B}{\hbar\omega}\frac{eE_0a}{\hbar\omega})\right|$ from the original gap $E_g^{(0)}$. Especially, when the argument of the Bessel function coincides with a zero point, the Peierls gap will collapse due to the coherent interaction with the laser field.

The dressed band for our model has been calculated by applying the Floquet theory. See Mizumoto and Kayanuma[16] for details of the calculation. In Fig. 2, examples of the calculated band-gap energy are shown against $eE_0a/\hbar\omega$ for two values of $\hbar\omega/E_g^{(0)}$. The agreement of the formula (14) (solid lines) with the numerical values (dashed lines) is good. See how the effect of the band-gap reduction is enhanced according to the increase of $B/\hbar\omega$.

The formula (14) reminds us of the formulas for CDT (Ref. 14) and the dynamic localization (DL).[12,17] However, there is an important difference between Eq. (14) and those for CDT and DL. In CDT and DL, the argument of the Bessel function is $eEa/\hbar\omega$, namely, the Stark energy between the neighboring atoms normalized by the energy quantum of the oscillating field. In order to effectively reduce the hopping energy, $eEa/\hbar\omega$ must be on the order of unity. For typical values $\hbar\omega \simeq 2$ eV and $a \simeq 0.1$ nm, this requires the amplitude of the laser field as high as $E_0 \simeq 2\times 10^{-10}$ V/m, or $I=4.8\times 10^{13}$ (W/cm^2). This makes it difficult to observe CDT or DL for electrons in crystals by using lasers in the optical region. Actually, the experimental observation of CDT and DL have been done in the cold atoms in the optical lattice.[18] In the collapse of band gap presented here, on the other hand, the effect of itinerancy of the conduction electron is incorporated in the factor $2B/\hbar\omega$ in the argument of the Bessel function. Since $2B/\hbar\omega \gg 1$, this additional factor makes it relatively easy to attain the condition for appreciable reduction of the band gap even for $eEa/\hbar\omega < 1$. For the band-gap renormalization in laser fields, Miranda[19] proposed a reduction in the gap under an intense field, based on the nearly free-electron model. It is an exercise to see that Miranda's result can be recovered as a special case of our model in the limit, $a \to 0$ and $B \to \infty$.

The analogy between the theoretical treatments to derive formulas for CDT and the band-gap collapse is still remarkable. In CDT, the oscillating field reduces the hopping amplitude *in real space*. It as been clarified that both of CDT and DL are regarded as a result of destructive interference in the repeated level crossings.[13] In the band-gap collapse, on the other hand, the oscillating field reduces the amplitude of resonant scattering *in the momentum space*. Both effects are thus understood as realizations of the coherent reduction in quantum transitions, in real space, and the momentum space, by a rapidly oscillating field.

So far, we have considered the stationary response of the electronic system to an applied monochromatic laser field for a fixed value of Q. Actual experiment will be done by a pump-probe measurement with a high-frequency short pump pulse. For the probe, a transient-induced absorption, or rather a transient photoemission spectroscopy will be desirable to observe the shift and broadening in the valence band. For the analysis, a theory of the time-dependent responses to such a short pulse should be formulated in future. Below we discuss only briefly some aspects to be expected. The equilibrium value Q_0 itself is determined self-consistently with the electronic energy as shown in Eq. (8). Therefore, the reduction in the effective coupling constant by the field will induce a motion of the lattice system. The dynamics of the total electron-lattice system should depend on the time scales of responses relative to the time scale of the temporal change in the pulse envelope τ_p. The response time of the electron τ_e is roughly estimated as $\tau_e \approx \hbar/E_g^{(0)}$ while that for the lattice $\tau_L \approx \omega_L^{-1}$ with ω_L being the frequency of the relevant phonon mode. The electronic system is expected to follow the rise of the pulse intensity adiabatically. If $\tau_p \gg \tau_L$, namely, if the rise of the envelope of the laser pulse is slow enough, the lattice system also follows the change in the electronic state adiabatically. On the contrary, if $\tau_p \ll \tau_L$, the sudden change in the electronic system will excite a violent motion of the lattice system around the new equilibrium point. Even in this case, there will be a time region shorter than τ_L in which the dynamical band-gap collapse is observed before the lattice motion begins.

One of the obstacles in the realization of the dynamical band-gap collapse proposed here may be the presence of real excitation of carriers, which hinders the observation of the coherent processes. One of the possibilities to overcome the difficulty will be going to the region of superintense field beyond the *Death Valley*,[20] where the higher-order coherent

process predominates over the lower-order process such as photoabsorption.

In the present work, it has been shown that an intense and high-frequency electromagnetic field may coherently suppress the quantum dynamics of electrons. If the period of the oscillative field is much faster than the time scale that dominates the quantum dynamics, the transitions of the electron may be frozen by the destructive interference.[13] As such, the mechanism of the intense field collapse is rather general and can be extended to other electronic interactions in matter. Details of the application to other systems will be a subject of future study.

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