

Peierls transition in a strong light field

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(Submitted March 11, 1976)

Pis'ma Zh. Eksp. Teor. Fiz. 23, No. 8, 449-451 (20 April 1976)

The influence exerted by a strong light field on the Peierls lattice instability is discussed. It is shown that the field induces a dielectric-metal transition.

PACS numbers: 71.40.+n

At the present time, in quasi-single-domain structures, such as charge-transfer salts based on tetracyanoquinodimethane (TCNQ) or plane-quadratic complexes of transition metals (Pt, Ir), have attracted attention in connection with the promise of realization of high temperature superconductivity in such systems.^[1] Without dwelling on all the aspects of the onset of the superconductivity in such metals, we point to one of the main factors, hindering the latter, namely the Peierls transition into the dielectric state at low temperatures. This raises the question of how to eliminate the Peierls transition by treating the crystal in one manner or another, i. e., how to metallize the system. From this point of view, we wish to discuss here the influence of a strong light field on a Peierls dielectric.

We emphasize that the presence of a narrow conductivity band $2|b| \approx 0.1-0.01$ eV in the substances indicated above, separated from the other bands by broad forbidden intervals, is very convenient from the point of view of using laser radiation with $\hbar\omega \sim 1$ eV, since it makes it possible to exclude the undesirable heating of the electrons by the light field. For a formal analysis of the influence of a strong light field on the Peierls transition, we consider the Fock-Hamiltonian^[1] \mathcal{H}_0 for a one-dimensional system of electrons, and supplement it with a term \mathcal{H}_{eff} responsible for the interaction of the electrons with the light wave.

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{eff}},$$

$$\mathcal{H}_0 = \sum_{\mathbf{k}} \epsilon_{\mathbf{k}} a_{\mathbf{k}}^{\dagger} a_{\mathbf{k}} + \sum_{\mathbf{q}} \hbar\omega_{\mathbf{q}} \beta_{\mathbf{q}}^{\dagger} \beta_{\mathbf{q}} + \sum_{\mathbf{k}, \mathbf{q}} D_{\mathbf{q}} a_{\mathbf{k}+\mathbf{q}}^{\dagger} a_{\mathbf{k}} \beta_{\mathbf{q}} + \text{H.c.}, \quad (1)$$

$$\mathcal{H}_{\text{eff}} = \sum_{\mathbf{k}} \lambda(\mathbf{k}) \sin\omega t a_{\mathbf{k}}^{\dagger} a_{\mathbf{k}}.$$

Here $a_{\mathbf{k}}^{\dagger}$, $a_{\mathbf{k}}$, $\beta_{\mathbf{q}}^{\dagger}$, and $\beta_{\mathbf{q}}$ are respectively the operators for the creation and annihilation of electrons with momentum \mathbf{k} and of phonons with momentum \mathbf{q} , $\epsilon_{\mathbf{k}} = \hbar v_{\mathbf{k}} \cos ka$ is the electron energy, a is the lattice constant, $\hbar\omega_{\mathbf{q}}$ is the phonon energy, $D_{\mathbf{q}}$ is the matrix element of the electron-phonon interaction, and $\lambda(\mathbf{k}) = \mathbf{v}_{\mathbf{k}} \cdot \sum_{\mathbf{q}} e/\omega$ is the matrix element of the intraband optical transitions in the field $\sum_{\mathbf{q}} \sin\omega t$ of the monochromatic light wave, and $\mathbf{v}_{\mathbf{k}}$ is the velocity of an electron with momentum \mathbf{k} .

We eliminate \mathcal{H}_{eff} from the Hamiltonian with the aid of the unitary transformation^[2]

$$U = \exp \left\{ \frac{i}{\hbar} \int_0^t \mathcal{H}_{ef} dr \right\}.$$

The transformed Hamiltonian reduces to the form

$$\mathcal{H} = \sum_{\mathbf{k}} \epsilon_{\mathbf{k}} a_{\mathbf{k}}^+ a_{\mathbf{k}} + \sum_{\mathbf{q}} \hbar \omega_{\mathbf{q}} \beta_{\mathbf{q}}^+ \beta_{\mathbf{q}} + \sum_{n \mathbf{k} \mathbf{q}} D_{\mathbf{q}} i^n J_n \left(\frac{\lambda(\mathbf{k} + \mathbf{q}) - \lambda(\mathbf{k})}{\hbar \omega} \right) e^{-in\omega t} a_{\mathbf{k} + \mathbf{q}}^+ a_{\mathbf{k}} \beta_{\mathbf{q}} + \text{H.c.}$$

Here $J_n(x)$ is a Bessel function with integer index.

We shall be interested henceforth in the case when the width in the gap in the electron spectrum is $\Delta \ll \hbar \omega$. Then, confining ourselves in (3) to the resonant terms ($n=0$), we ultimately have

$$\mathcal{H} = \sum_{\mathbf{k}} \epsilon_{\mathbf{k}} a_{\mathbf{k}}^+ a_{\mathbf{k}} + \sum_{\mathbf{q}} \hbar \omega_{\mathbf{q}} \beta_{\mathbf{q}}^+ \beta_{\mathbf{q}} + \sum_{n \mathbf{k} \mathbf{q}} D_{\mathbf{q}} J_0 \left(\frac{\lambda(\mathbf{k} + \mathbf{q}) - \lambda(\mathbf{k})}{\hbar \omega} \right) a_{\mathbf{k} + \mathbf{q}}^+ a_{\mathbf{k}} \beta_{\mathbf{q}} + \text{H.c.} \quad (4)$$

The Hamiltonian (4) differs from the Fröhlich Hamiltonian in the absence of a light wave by a renormalization of the constant $D_{\mathbf{q}}$:

$$\tilde{D}_{\mathbf{q}} = D_{\mathbf{q}} J_0 \left(\frac{\lambda(\mathbf{k} + \mathbf{q}) - \lambda(\mathbf{k})}{\hbar \omega} \right),$$

which now becomes dependent on the intensity of the light field. We can therefore write down directly an expression for the gap in the Peierls-dielectric spectrum at $T=0$ ^[1,3]

$$\Delta = 8 |b| e^{-1/g}, \quad (5)$$

where

$$\tilde{g} = g J_0 \left(\frac{\lambda(\mathbf{k}_F) - \lambda(-\mathbf{k}_F)}{\hbar \omega} \right),$$

g is the interaction constant in the absence of the field, and k_F is the momentum on the Fermi surface. Accordingly, the critical temperature is

$$T_p = \frac{\gamma}{\pi} 8 |b| e^{-1/g}, \quad (6)$$

where $\ln \gamma = C$ is the Euler constant.

It follows from (5) that the gap Δ , as well as T_p , will oscillate with increasing light field, vanishing at the zeroes of the Bessel function $J_0(x)$. Thus, the light field induces a dielectric-metal transition.

The appearance in the crystal spectrum of the gap under the Peierls instability can be interpreted as a consequence of the onset of an additional period π/k_S in the crystal. The vibrational motion of the electron in the field of the light wave causes the action of the additional periodic potential on the lattice to become averaged over the period of the oscillations. The averaged potential vanishes in fields in which the doubled oscillation amplitude of the electron $2e \mathcal{E}/m\omega^2$ becomes equal to or a multiple of the period π/k_F , and this leads to oscillations of the gap.

Let us estimate the minimum value of the field, at which $\Delta=0$: $\vec{\mathcal{E}}$ is parallel to the conducting filaments. Approximating $\epsilon_{\mathbf{k}} = b \cos ka$, putting $2|b| \approx 0.1$ eV, $\hbar \omega \approx 0.14$ eV (CO₂ laser), and $a \approx 4$ Å, and recognizing that $J_0(x) = 0$ at $x = 2.4$, we have

$$\mathcal{E} = 2.4 \frac{\hbar^2 \omega^2}{2|b|ae} \approx 8 \times 10^6 \text{ V/cm.} \quad (7)$$

As seen from the estimate (7), the light fields at which the gap vanishes completely at $T=0$ are close to their breakdown values. However, for practical purposes, such as the search for high-temperature superconductivity, it suffices only to shift T_p somewhat into the region of lower temperatures, which is possible in much weaker fields. (A shift of T_p by 1 °K corresponds to fields 10^4 – 10^5 V/cm, and accordingly to light fluxes ~ 0.1 – 10 MW/cm²).

In conclusion, we indicate clearly qualitatively one more possibility of controlling the Peierls transition by using a strong light field. It is known that the temperature of the Peierls transition depends strongly on the degree of one-dimensionality of the system, which is characterized by the ratio $\sigma_{||}/\sigma_{\perp}$ of the conductivity along the filaments to the hopping conductivity in the perpendicular direction. It is also known that a light field that is almost at resonance with the energy interval between the conduction band and the unfilled bands that lie above it should lead to an increase of σ_{\perp} . Thus, by illuminating the crystal with radiation of varying power we are able to regulate the degree of one-dimensionality of the system, and choose conditions optimal for the observation of the superconductivity.

The author is grateful to S. B. Beneslavskiĭ and L. N. Bulaevskiĭ for a useful discussion of the work.

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