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QUASIENERGY BAND STRUCTURE OF SOLIDS

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The structure of the energy band spectrum of a solid in an external laser field presents a rather interesting and challenging problem.[1-5] On one hand the spatial periodicity of the solid leads to the conventional band theory of solids. The one electron states are characterized by the crystal momentum whose value is restricted to the first Brillouin zone (FBZ), and a band index (as well as a spin index). On the other hand the temporal periodicity imposed by the laser field combined with a Floquet analysis leads one to a picture in which one need only consider the band structure in the first omega zone (FOZ), i.e. in the range $0 \le \epsilon \le \hbar\omega$, where ω is the laser frequency. In such a system energy is no longer a good quantum number, and one is naturally led to investigation of the quasinergy bands defined in the hyperrectangle $k \in$ FBZ, $\epsilon \in$ FOZ. However, surprisingly few papers have been dedicated to such an investigation,[2,3] and with such a point of view. Generally, the interest in temporally periodic Hamiltonian problems has been confined to atomic systems,[4] or eventually to two-band solid models.[5] It is the main purpose of this communication to turn attention to the multiply periodic solid-in-laser problem, stressing the alternative, but equally legitimate approach via quasienergy bands.

Basic questions remain to be answered regarding this problem. What is the mathematical nature of the spectrum? Since the band structure problem has been reduced to the knowledge of the spectrum in the hyperrectangle, how are the eigenvalues distributed? Are they dense, i.e. do they completely fill the hyperrectangle or do gaps exist? Are there discrete eigenvalues? A rigorous answer to these questions is difficult to obtain, and may be dependent on the model or approximations used. So, we set out here on a more modest course, by first identifying the difficulties of the general case, and then treating a simple model which yields some useful results.

The system of interest is a crystalline solid interacting with an electromagnetic field. The one-electron Hamiltonian in the $\mathbf{A}\cdot\mathbf{p}$ gauge and MKS units is of the form:

$$H(\mathbf{p},\mathbf{r},t) = \frac{p^2}{2M} + V(\mathbf{r}) - \frac{e}{M}\mathbf{A}\cdot\mathbf{p} , \tag{1}$$

where $V(\mathbf{r})=V(\mathbf{r}+\mathbf{R})$ is the crystal potential ($\mathbf{R}$ being any lattice vector), and e and M are the electronic charge and mass respectively. We will consider the special case of a space-homogeneous time-periodic vector potential $\mathbf{A}(t)=\mathbf{A}(t+\tau)$. In this case the problem of solving the time-dependent Schrödinger equation:

$$i\hbar\, \partial_t | \psi_t > = H(\mathbf{p},\mathbf{r},t) | \psi_t > \tag{2}$$

is simplified by the use of the Floquet theory,[2-4] according to which the state vector may be chosen as:

$$| \psi_t > = \exp(-i\epsilon t) \, | u_t > , \tag{3}$$

with $|u_t> = |u_{t+\tau}>$ being time-periodic. Upon performing the Fourier transformation an infinite set of operator equations is obtained:

$$\left[\frac{p^2}{2M}+V(\mathbf{r})-\hbar(\epsilon+n\omega)\right] | u_n > = \frac{e}{M}\mathbf{p}\cdot\sum_{m=-\infty}^{\infty}\mathbf{A}_{n-m} | u_m >, \tag{4}$$

where:

$$| u_n > = \frac{1}{2\pi}\int_0^{2\pi}\exp(in\omega t) \, | u_t > d\omega t , \tag{5}$$

$$\mathbf{A}_n = \frac{1}{2\pi}\int_0^{2\pi}\exp(in\omega t)\, \mathbf{A}(t)\, d\omega t , \tag{5a}$$

are the temporal Fourier components, $\omega\tau=2\pi$, and n can go from $-\infty$ to $+\infty$. Let us call $H_0(\mathbf{p},\mathbf{r}) = \frac{p^2}{2M} + V(\mathbf{r})$, the ideal crystal Hamiltonian. One way to proceed is to go to the Schrödinger picture:

$$[H_0(-i\hbar\nabla,\mathbf{r})-\hbar(\epsilon+n\omega)]u_n(\mathbf{r}) = \frac{e}{M}(-i\hbar\nabla)\cdot\sum_m \mathbf{A}_{n-m}u_m(\mathbf{r}) , \tag{6}$$

and then perform the Bloch analysis, i.e. pick the solution in the form:

$$u_n(\mathbf{r}) = \exp(i\mathbf{k}\cdot\mathbf{r})\, v_n(\mathbf{r}) , \tag{7}$$

where $v_n(\mathbf{r}+\mathbf{R})=v_n(\mathbf{r})$, and then Fourier analyse in $\mathbf{r}$:

$$v_n(\mathbf{r}) = \sum_{\mathbf{K}}\exp(i\mathbf{K}\cdot\mathbf{r})\, v_{n\mathbf{K}} , \tag{8a}$$

$$V(\mathbf{r}) = \sum_{\mathbf{K}}\exp(i\mathbf{K}\cdot\mathbf{r})\, V_{\mathbf{K}} , \tag{8b}$$

where **K** is any reciprocal lattice vector. The result is a doubly infinite system:

$$\left[\frac{\hbar^2}{2M}(\mathbf{k}+\mathbf{K})^2 - \hbar(\epsilon+n\omega)\right]v_{n\mathbf{K}} + \sum_{\mathbf{K}'} V_{\mathbf{K}-\mathbf{K}'}v_{n\mathbf{K}'} = \frac{e\hbar}{M}(\mathbf{k}+\mathbf{K})\cdot\sum_m \mathbf{A}_{n-m}v_{m\mathbf{K}}\ , \quad (9)$$

and the zeros of the Hill determinant

$$\Delta(\mathbf{k},\epsilon) = det\{\delta_{\mathbf{K},\mathbf{K}'}\,\delta_{n,n} + \frac{1}{\frac{\hbar^2}{2M}(\mathbf{k}+\mathbf{K})^2-\hbar(\epsilon+n\omega)}\left[V_{\mathbf{K}-\mathbf{K}'}\delta_{n,n'} - \frac{e\hbar}{M}(\mathbf{k}+\mathbf{K})\mathbf{A}_{n-n'}\delta_{\mathbf{K},\mathbf{K}'}\right]\} \quad (10)$$

determine the eigenvalues $\epsilon_\nu(\mathbf{k},\omega)$. The subscript ν labels the spectrum: it may take descrete or continuous values. Although the form of $\Delta(\mathbf{k},\epsilon)$ is not convenient for calculations, some important properties of the spectrum follow from it.[5]

First, the Hill determinant is invariant under translation of **k** by any reciprocal lattice vector:

$$\Delta(\mathbf{k}+\mathbf{K},\epsilon) = \Delta(\mathbf{k},\epsilon)\ . \quad (11a)$$

In this respect the band structure problem is similar to the ordinary solid-state theory: ones attention can be restricted only to the first Brillouin zone. However, the Hill determinant is also invariant under translation of ϵ by any number of ω:

$$\Delta(\mathbf{k},\epsilon+n\omega) = \Delta(\mathbf{k},\epsilon)\ . \quad (11b)$$

Thus, the region of interest in ϵ can also be restricted to the first ω zone. The band structure problem of a solid in a strong laser is confined to a fundamental hyperrectangle in the ϵ-**k** space.

The other way to proceed in addressing this problem is to project the equations (4) on the Bloch states of the ideal solid $\langle \mathbf{r}|n'\mathbf{k}\rangle = \exp(i\mathbf{k}\mathbf{r})\ \omega_{n'}(\mathbf{r})$:

$$[E_{n'}(\mathbf{k})-\hbar(\epsilon+n\omega)]\langle n'\mathbf{k}|u_n\rangle = \frac{e\hbar}{M}\sum_m \mathbf{k}\mathbf{A}_{n-m}\langle n'\mathbf{k}|u_m\rangle - \frac{ie\hbar}{M}\sum_m\sum_{n''}\int d^3r\ \omega_{n'}^*\ \nabla\omega_{n''}\cdot\mathbf{A}_{n-m}\langle n''\mathbf{k}|u_m\rangle \quad (12)$$

where $E_{n'}(\mathbf{k})$ is the band energy of the unperturbed solid, and the integral is taken over the unit cell. Thus, the only way in which energy bands couple is via the momentum matrix elements,

$$\langle n'\mathbf{k}'|\mathbf{p}|n''\mathbf{k}''\rangle = \delta(\mathbf{k}'-\mathbf{k}'')\ \mathbf{p}_{n',n''} = \delta(\mathbf{k}'-\mathbf{k}'')\left[\hbar\mathbf{k}'\ \delta_{n',n''} - i\hbar\int d^3r\ \omega_{n'}(\mathbf{r})\ \nabla\omega_{n''}(\mathbf{r})\right]\ . \quad (13)$$

As it stands, the system of coupled equations (12) cannot be solved analytically. Its numerical treatment also entails considerable difficulties. That place is usually a branching point for any research. From there on usually more specific aspects of the problem are addressed and various approximations introduced - like the two-band models, a monochromatic laser, various approximations for Bloch states, etc.[1-5] *A* common reason is self-evident intractability of the general problem.

Since our goal is to bring attention to the concept of quasienergy bands, and not to make detailed calculations, we will accordingly restrict ourselves to a simple effective mass nearly-free electron model.[1] In the lowest order it consists in approximating the momentum by the quasimomentum. In doing so, however, we should be aware of the familiar danger in theoretical work: the general theory with interesting phenomena may be intractable, and the tractable models may be uninteresting. With this in mind, let us evaluate the quasienergy spectrum of the effective mass model.

The system (12) significantly simplifies in this approximation. Since the quasimomentum is diagonal in the crystal momentum representation, no more momentum-mixing of energy bands occurs:

$$[E_{n'}(\mathbf{k})-\hbar(\epsilon+n\omega)]\langle n'\mathbf{k}|u_n\rangle = \mathbf{k}\cdot\frac{e\hbar}{\mathbf{M}^*}\cdot\sum_m \mathbf{A}_{n-m}\langle n'\mathbf{k}|u_m\rangle, \quad (14)$$

where $1/\mathbf{M}^*$ is the reciprocal of the effective mass tensor. The no-mixing feature sweeps under the rug many of the problems and controversies[6] surrounding the subject. The quasienergy $\epsilon(\mathbf{k},\omega)$ in the effective mass approximation is given by the solution of:

$$\Delta(\mathbf{k},\epsilon) = det\left[\delta_{m,n} - \frac{\mathbf{k}\cdot\frac{e\hbar}{\mathbf{M}^*}\cdot\mathbf{A}_{n-m}}{E_{n'}(\mathbf{k})-\hbar(\epsilon+n\omega)}\right] = 0\ . \quad (15a)$$

We will also consider only the principal mode of the laser $\mathbf{A}=\mathbf{A}_0\cos\,\omega t$, without much further loss of generality. Hence $\mathbf{A}_{n-m} = \mathbf{A}_0(\delta_{m,n+1} + \delta_{m,n-1})/2$, and:

$$\Delta = det\left[\delta_{m,n} - \frac{a}{E_{n'}(\mathbf{k})-\hbar(\epsilon+n\omega)}(\delta_{m,n-1}+\delta_{m,n+1})\right], \quad (15b)$$

where $a=\mathbf{k}\cdot\frac{e\hbar}{2\mathbf{M}^*}\cdot\mathbf{A}_0$. This infinite tridiagonal determinant, or a continuant,[7] is absolutely convergent, since:

$$\sum_m \frac{a}{E_{n'}(\mathbf{k})-\hbar(\epsilon+m\omega)}\,\frac{a}{E_{n'}(\mathbf{k})-\hbar(\epsilon+m\omega+\omega)} = \frac{\pi a^2}{(\hbar\omega)^2}\left[\cot\frac{\pi}{\hbar\omega}(E_{n'}-\hbar\epsilon) - \cot\frac{\pi}{\hbar\omega}(E_{n'}-\hbar(\epsilon+\omega))\right] = 0\ . \quad (16)$$

As it is readily seen, $\Delta(\mathbf{k},\epsilon)$ is periodic in ϵ, and has simple poles at $E_{n'}(\mathbf{k})-\hbar(\epsilon+m\omega) = 0$. Furthermore, it is bounded at infinity $[\Delta(\mathbf{k},\infty)=1]$, so it may be written in the form:[7,8]

$$\Delta(\mathbf{k},\epsilon) = 1 + C\sum_m \frac{a}{E_{n'}(\mathbf{k})-\hbar(\epsilon+m\omega)}\ , \quad (17a)$$

where C is a constant, proportional to the residue at each of the poles of Δ. After performing the summation:

$$\Delta = 1 + \frac{Ca\pi}{\hbar\omega}\cot\frac{\pi}{\hbar\omega}[E_{n'}(\mathbf{k})-\hbar\epsilon]\ , \quad (17b)$$

so that $C=\frac{\hbar\omega[\Delta(\mathbf{k},0)-1]}{a\pi\cot\pi/\hbar\omega\ E_{n'}(\mathbf{k})}$, and the quasienergy is implicitly given by:

$$\cot\frac{\pi}{\hbar\omega}[\hbar\epsilon - E_{n'}(\mathbf{k})] = \frac{\cot\frac{\pi}{\hbar\omega}E_{n'}(\mathbf{k})}{\Delta(\mathbf{k},0) - 1}\ . \quad (18)$$

In this case the quasienergy can be labeled by the same band index n' (but the actual order of the bands will be different), indicating that not much interesting is happening in this model. It is not good for strong fields, and applicable only around extremal points in the k-space; its utility is restricted to a pictorial introduction.

A more interesting insight into the quasienergy band structure is provided by considering the zero-field limit of the general theory, and then slowly turning the field on. In the zero-field case the quasienergy band structure is obtained by slicing the ordinary energy bands at different $n\omega$ values, and bringing the pieces down to the first ω zone. Few of the typical situations which may arise are depicted in figure 1. As it is evident, the pieces cross each other at many points, and the density of crossing points is not uniform. The number of crossings is at most denumerable. In principle, as more and more segments are brought down, it is conceivable that they will cover the whole hyperrectangle fairly uniformly, producing a dense spectrum. In other words, an electron can find itself arbitrarily close to any point in the hyperrectangle.

In practice, however the number of segments that need be translated down to FOZ for a realistic description is finite, limited by such considerations as the position of the Fermi level in the unperturbed solid and the intensity of the oncoming laser, which restricts the order of multiphoton processess coupling widely separated segments. The number of translations though, increases with the intensity.

As the interaction is turned on, gaps open at the crossings and the bands shift. In the limit of high field intensity it is not clear in which way the gaps change appearence of the quasispectrum. It seems unreasonable that they would eject band lines from any wide area of the hyperrectangle. The size of a gap depends on the order of the process (i.e. on the number of downward translations): one-photon processess (one translation) produce larger gaps. The gaps also widen as the field gets stronger. But the field effect on the already existing gaps at the edges of FBZ is exactly the opposite:[9] for low fields it reduces the size of the gap.

In the weak perturbation limit the size of a gap can be estimated directly from the Hill determinant following from Eq. (12):

$$\Delta = det\left[\delta_{n',m'}\delta_{n,m} - \frac{\frac{e}{M}\,\mathbf{p}_{n',m'}\cdot\mathbf{A}_{n-m}}{E_{n'}-\hbar\omega(\epsilon+n\omega)}\right]. \tag{19}$$

For weak fields:

$$\Delta = 1 - \frac{1}{2}\sum_{n,n',m,m'} \frac{(e/M)^2\left|\mathbf{p}_{n',m'}\cdot\mathbf{A}_{n-m}\right|^2}{[E_{n'}-\hbar(\epsilon+n\omega)][E_{m'}-\hbar(\epsilon+m\omega)]}, \tag{20}$$

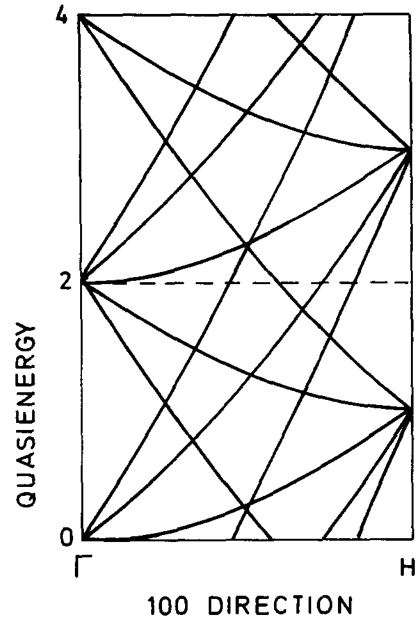
and near the intersection both denominators are small for some set of integers n,n',m,m'. The two quasienergy bands are determined by solving

$$[E_{n'}-\hbar(\epsilon+n\omega)][E_{m'}-\hbar(\epsilon+m\omega)]=(e/M)^2\left|\mathbf{p}_{n',m'}\cdot\mathbf{A}_{n-m}\right|^2, \tag{21}$$

and the size of the gap is given by:

$$E_g = 2\,(e/M)\,\left|\mathbf{p}_{n',m'}\cdot\mathbf{A}_{n-m}\right|. \tag{22}$$

In conclusion, the aim of this communication is not to present detailed and definitive results concerning alterations in the band structure of solids under the influence of intense laser light, but to stimulate thinking in the direction of the usually overlooked concept of quasienergy bands.



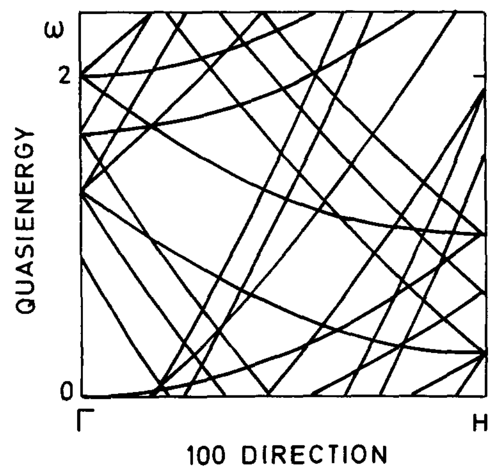


Figure 1(*a*): Quasienergy bands of a BCC lattice in the nearly-free electron approximation, in the 100 direction (parallel to the field). Energy unit is $\frac{(\pi\hbar)^2}{2Ma^2}$, and the frequency ω is chosen so that $\hbar\omega$=2, a highly degenerate case. Only the first twelve unperturbed bands are taken into account, which are seen to collapse to five quasienergy bands. Two omega zones are plotted, to display periodicity.
(*b*): Same as (*a*), except that $\hbar\omega$ is slightly larger. The bands do not close on themselves as in the comensurate case of (*a*). All 12 bands are now visible.

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