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Electron localization in crystals with quasiperiodic lattice potentials

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Anderson's locator perturbation theory, used in the study of localization in disordered systems, is applied to the study of electron localization in tight-binding model lattices containing a periodic modulation potential incommensurate with the crystal lattice. Numerical studies of the convergence of the resulting continued fraction for the self-energy indicate that in one dimension there is a transition at a critical value of strength of the modulation from all states localized to all states extended, unlike the disordered crystal case. Studies in two and three dimensions show that there exists an intermediate range of modulation strength over which there can be mobility edges separating localized and extended states. Specific application is made of the results in one dimension to a system containing a superlattice modulation in one direction.

I. INTRODUCTION

Recently, Azbel[1] has argued that the spectrum of an electron moving in periodic potentials with incommensurate periods is of the "devil's-stair" type, with both localized and extended states separated by mobility edges. On the other hand, Aubry[2] has argued, for a tight-binding one-dimensional model with a sinusoidal potential incommensurate with the lattice period, that the electronic states would be all localized for the sinusoidal potential strength greater than a critical value and all Bloch-type extended states for smaller values of this potential, with no mobility edges. The present author argued[3] that the phonons in Aubry's model are localized in wave-vector space for sufficiently weak sinusoidal potential strength, implying that the phonons are not damped, despite the fact that such a system lacks translational symmetry. The arguments used in that paper were based on Anderson's locator expansion, which is used in discussing localization in the disordered-crystal problem.[4,5,6] If the arguments of this paper are applied in r space, they imply that for sinusoidal potential stronger than a critical strength all states are localized in r space, in agreement with Aubry.

Reference 3, however, instead of using the renormalized perturbation theory, replaced the mass-operator perturbation theory by a geometric series, each term of which was the geometric average of each term in the actual perturbation theory. This is certainly not a valid procedure in the limit in which the period of the sinusoidal potential is much larger than a lattice constant, the limit considered by Azbel. The reason for this is that in such a limit the energy denominators, associated with neighboring sites in the perturbation theory, are nearly equal. Therefore, processes in which an electron moves back and forth many times between neighboring sites will cause the self-energy perturbation theory to diverge. Such divergences were discussed in Refs. 4, 5, and 6. They do not imply the existence of a branch cut in the self-energy, and they can be eliminated by the Watson renormalization trick.[4,5,6,7] In this article, the Watson procedure is applied to this problem. In one dimension, the renormalized perturbation theory for the self-energy leads to a pair of infinite continued fractions.[6] The convergence of these continued fractions will be studied numerically, and the results will be compared with Azbel's and Aubry's results. The advantage of the locator perturbation method over the method of Azbel or Aubry is that it can be applied in higher dimensions. In Sec. II, the tight-binding model will be formulated, and it will be discussed in the light of Azbel's theory. In Sec. III, convergence of the continued fractions for the self-energy will be discussed and Azbel's and Aubry's results will be discussed in the light of these results. An extension of the present results to two and three dimensions will also be presented. In Sec. IV, application of these results to real experimental systems will be discussed.

II. THE TIGHT-BINDING MODEL

The model studied in this paper is a tight-binding model whose Schrödinger equation is

$$f_{n+1}+f_{n-1}+V_0(\cos qn)f_n=Ef_n\,, \tag{1}$$

where f_n is the expansion coefficient for the tight-binding function

$$\psi(x)=\sum_n f_n\varphi(x-na)\,, \tag{2}$$

where φ is an atomic or Wannier function. We will be interested in the case in which q is an irrational multiple of π. The dimensionless energy and potential strength are assumed to be measured in units of the intersite hopping matrix element.

Aubry[2] makes use of the transformation to a reciprocal-lattice representation,

$$f_n = \sum_m g_m e^{i(k+qm)n}, \tag{3}$$

which transforms Eq. (1) to

$$g_{m+1} + g_{m-1} + V_0' \cos(qm + k) g_m = E' g_m, \tag{4}$$

where $E' = 2E/V_0$ and $V_0' = 4/V_0$. By using a formula for the exponential decay of a wave function due to Thouless, Aubrey[2] shows that all solutions to Eq. (1) are localized for $V_0 > 2$, and all solutions to Eq. (4) are obviously localized if $V_0' > 2$ or $V_0 < 2$.

Azbel[1] discusses the Kronig-Penny model, but his ideas can be applied to the present model. To do so we define a position variable $r = qn$ and a momentum operator $\hat{p} = iq(\partial/\partial r)$ and obtain a Schrödinger equation

$$2\cos(\hat{p}) f_r + V_0 \cos(r) f_r = E f_r \tag{5}$$

whose "classical" (i.e., $q \to 0$) trajectory is given by

$$E = 2\cos p + V_0 \cos(r), \tag{6}$$

where p is a c number. It is easy to show that for $V_0 > 2$, all trajectories are localized orbits (in r). For $V_0 < 2$ there exist only localized orbits for $|E| > |2 - V_0|$ and only extended orbits for $|E| < |2 - V_0|$, in the sense that an electron in a classically localized orbit moves in a finite closed orbit and an electron in an extended orbit travels over the whole crystal. Azbel's analysis would show that for small but finite q the localized orbits are quantized and the states near the mobility edge[1] can become broadened into narrow bands, but the localized classical orbits far from the mobility edge stay pretty much localized. Of course, even the extended states must possess, for any finite q, a gap at each energy, although most of the steps will be infinitesimally small. Most likely all but the largest band gaps will be so small that Zener breakdown will occur for even extremely weak fields. In this sense, such states may be considered extended and able to conduct electricity.[1] Thus, we conclude that Azbel's theory predicts for the present tight-binding model the occurrence of mobility edges near $\pm(2 - V_0)$.

III. LOCATOR EXPANSION THEORY OF LOCALIZATION IN INCOMMENSURATE SYSTEMS

The condition that the solutions to Eq. (1) be localized is equivalent to requiring that the perturbation theory for the self-energy $M(E)$ of the Green's function for Eq. (1) converge for real energies, implying that the self-energy does not have a branch cut on the real energy axis.[4,5] In one dimension, when the Watson renormalized procedure[6] is applied to this perturbation series, the self-energy can be expressed in the standard form,[6,8]

$$M(E) = \cfrac{2}{a_1 + \cfrac{1}{a_1 + \cfrac{1}{a_2 + \cfrac{1}{a_3 + \cfrac{1}{a_4 + \cfrac{1}{a_5 + \cdots}}}}}}, \tag{7}$$

where

$$a_n = (V_0 \cos nq - E)(-1)^n. \tag{8}$$

If we cut off the continued fraction after n terms, it may be written as a ratio of polynomials

$$M(E) = 2(P_n/Q_n), \tag{9}$$

where P_n and Q_n satisfy the recursion relation,[8]

$$P_n = a_n P_{n-1} + P_{n-2}, \tag{10}$$

$$Q_n = a_n Q_{n-1} + Q_{n-2}, \tag{11}$$

where $P_1 = 1$ and $Q_1 = a_1$. We say that $M(E)$ converges if

$$\lim_{n\to\infty} \frac{P_n}{Q_n}$$

exists. It can be shown that[9]

$$\left| \frac{P_n}{Q_n} - \frac{P_{n-1}}{Q_{n-1}} \right| \leq \frac{1}{|Q_n Q_{n-1}|}, \tag{12}$$

implying that the continued fraction converges if $D = |Q_n Q_{n-1}| \to \infty$ as n approaches infinity. Convergence of Eq. (7) was studied using this condition and numerically iterating Eq. (11). We have iterated Eq. (11) up to $n = 1000$ and, in questionable cases, as high as $n = 10\,000$, usually stopping the iterations when D reached 1000 or more, but sometimes letting it reach 10 000. In order to be sure that the results were free of roundoff error, runs were made with double and quadruple precision, and the results were compared. No sign of roundoff error problems was detected.

In Table I we see the results of such runs for both commensurate and incommensurate values of q, for various values of V_0. We see that for $V_0 > 2$, all states are localized for the incommensurate but not for the commensurate values. To illustrate the difference between the results for commensurate and incommensurate q, consider $q = \pi$. For this commensurate case, Eq. (7) reduces to

TABLE I. Denominators for continued fraction for r space self-energy for an 1100-term (or fewer) continued fraction.

q	V_0	E	Q_nQ_{n-1}	Comment
3.141 592 7	2.1	2.1	1100	Localized
		2.2	1.231 588	Extended
		2.3	1.000 876 2	Extended
		2.4	0.234 062 42	Extended
		2.5	0.123 734 828	Extended
		2.6	$5.667\,279\,34\times10^{-2}$	Extended
3.1	2.1	1.8	1110	Localized
		1.9	1141	Localized
		1.95	1330	Localized
		2.0	1240	Localized
		2.1	1560	Localized
		2.2	2232	Localized
		2.3	2240	Localized
		2.4	1302	Localized
		2.5	1689	Localized
	1.9	1.89	1367	Localized
		1.9	7.956 89	Extended
		1.95	2.558 672	Extended
		2.0	1166	Localized
		2.1	1214	Localized
				All remaining are localized

$$M=2\bar{M},$$

where

$$\bar{M}=\frac{1}{E+V_0-1/E-V_0-\bar{M}}, \tag{13}$$

or

$$(E+V_0)\bar{M}^2-(E^2-V_0^2)\bar{M}+E-V_0=0.$$

We see from the solution to this equation that $\bar{M}$ possesses a branch cut in the range

$$V_0^2<E^2<V_0^2+4; \tag{14}$$

these are the band energies, and the remainder of the energies fall in band gaps. For $V_0>2.1$, we see from Table I that there are no extended states for the incommensurate-q case close to $q=\pi$. For V_0 less than 2, there do appear to be bands, however. At this point there is one point which must be clarified in this procedure. When Eq. (7) diverges, we know that there are extended states. When Eq. (7) converges, we could either have a localized state or a gap at that energy. To determine whether or not we have a state in an energy range for which Eq. (7) converges, we must examine the poles of the Green's function in wave-vector space [i.e., Eq. (4)]. To do this, we calculate the energy denominator

$$D=E-V_0'\cos(k+qm)-M'(k,E)$$

as a function of energy E for several wave vectors. If D passes through zero at a certain value of E, we conclude that there is a state at that energy; if D has no zeros, we are in a gap. It should be noted in applying this procedure that a change in sign of D does not necessarily imply a zero at an intermediate energy; it might instead imply that D passes through infinity. Of course, at each energy and wave vector one must check to see that the continued fraction used to calculate $M'(k,E)$ actually converged. Lack of convergence implies a branch cut at that wave vector, which certainly implies that there exist states at the energy in question. For $V_0=1.9$, states were found in this way at discrete energies between $E=0$ and 1.84, and for $E>2.0$. Thus we conclude that there are localized states, and hence mobility edges for $V_0<2$.

Let us now consider the low-q limit (i.e., Azbel's "classical" limit[1]). Studies of the convergence of the self-energy for $q=0.1$ and $V_0=1.0$ are given in Table II. A mobility edge is found at $E=1.0$ in agreement with the predictions of the "classical trajectories" discussed in Sec. II. Other such studies with different values of V_0 confirm the fact that a mobility edge appears to occur at $|E|=|2-V_0|$ as predicted in Sec. II. In Table II, the same study is also made for $q=0.1$ and $V_0=0.005$. The energies $E>1.996$ and near 1.99 (the regions where the real space self-energy converged) were tested to see whether there existed states at these energies or whether the energies

TABLE II. Denominators for continued fraction for r space self-energy for a 1100-term (or fewer) continued fraction. (* signifies a 10 100-term continued fraction.)

q	V_0	E	Q_nQ_{n-1}	Comment
0.1	1.0	0	0.354 030 7	Extended
		0.3	0.509 039 2	Extended
		0.5	5.832 434 5	Extended
		0.7	0.302 891 87	Extended
		1.0	0.296 03	Extended
		1.1	1434	Localized
		1.3	2517	Localized
		1.6	1284	Localized
		1.8	3586	Localized
		2	2916	Localized
		2.2	1665	Localized
0.1	0.005	0	$1.267\,931\times10^{-2}$	Extended
		0.2	0.335 791 39	Extended
		0.5	$4.792\,936\,06\times10^{-2}$	Extended
		0.7	0.588 492 6	Extended
		0.9	0.796 613 3	Extended
		1.0	0.995 106	Extended
		1.2	0.749 582 98	Extended
		1.5	0.249 959	Extended
		1.8	4.140 527 7	Extended
		1.9	0.775 996	Extended
		1.98	8.579 779 39	Extended
		1.988	55.273 8	Extended
		1.989	*93.605 5	Extended
		1.99	1131	Localized
		1.995	5.062 358 4	Extended
		1.996	1102	Localized
		2.00	1233	Localized

were in gap regions. States were found at discrete energies between 1.996 and 2.0, but none were found around $E=1.99$, indicating an energy gap. Similar studies were made with other small values of q and V_0 which also seem to indicate that there are localized states and bands of extended states separated by gaps. This implies that the mobility edge that occurs in the small-q limit is due to low-order energy gaps coalescing near the band edges as $q\to0$, giving rise to many very flat bands separated by many small-gap regions in the small-q limit. The higher-order gaps, which occur away from the band edges rapidly decrease in size with increasing order, becoming negligible in the "classically accessible region." The implication is that it appears as if, for small but nonzero q, there does not exist a mobility edge. Rather, we get a collection of extremely narrow bands in the "classically inaccessible region."

The incommensurate system described in this article, like a disordered system, has no translational order. Unlike the disordered system, however, for which all states are localized in one dimension, the incommensurate system appears to have a sharp transition from a phase in which all electric states are localized to one in which most states are extended, as a function of the strength of the second periodic potential.

One of the advantages of the formulation of the incommensurate lattice problem in terms of locator perturbation theory used in the Anderson localization problem is that the same method can be applied in two and three dimensions. Consider the following two- or three-dimensional generalization of the equation of motion [i.e., Eq. (1)]:

$$\Sigma_{\vec{a}} f(\vec{R}+\vec{a}) + v(\vec{R}) f(\vec{R}) = Ef(\vec{R}), \tag{15}$$

where $\vec{R}$ is any lattice vector, $\vec{a}$ is a near-neighbor lattice vector, and $v(\vec{R})$ is a periodic potential incommensurate with the lattice (i.e., having no reciprocal-lattice vectors in common with the lattice). Then, the perturbation theory for the self-energy of the Green's function of Eq. (15) can be written, using standard methods,[5] as

$$M(\vec{R},E)=\Sigma_n\Sigma_{\vec{a}_1\vec{a}_2,\ldots,\vec{a}_n}\frac{1}{E-v(\vec{R}+\vec{a}_1)-M_1}\times\frac{1}{E-v(\vec{R}+\vec{a}_1+\vec{a}_2)-M_{1,2}}\cdots, \tag{16}$$

where included in the sum are only self-avoiding random walks starting and ending on site R, and where the $M_{1,2,3,\ldots,n}$ have the usual meaning of being the sum of paths returning to site n without passing sites $1,2,\ldots,n$ at any intermediate point along the way. Because of the incommensurate nature of the system, no two denominators will be equal.[1] Thus for almost all energies, there will be no divergences of the perturbation theory for a finite system; divergences only occur for an infinite system for which there are infinitely many terms in the series. The possibility of divergence of Eq. (16) for an infinitely large system may be tested by examining the convergence of the geometric series whose ratio is the geometric mean of the reciprocal of an energy denominator in Eq. (16).[4,5] As is the usual procedure, the self-energies of the energy denominators of Eq. (16), assumed to be well behaved and not having an effect on the convergence of Eq. (16), are neglected.[4,5] Then the geometric mean of an energy denominator $D(E)$ is defined by

$$\ln D(E)=\frac{1}{\Omega}\int d^dR \ln|E-v(\vec{R})|, \tag{17}$$

where the integral is taken over a volume Ω ($\vec{R}$ is treated as a continuous variable), where Ω is the unit cell of potential $v(\vec{R})$, i.e., the smallest volume over which $v(\vec{R})$ takes on all its possible values once. Then, the geometric series that we wish to examine is

$$\Sigma_n [KD^{-1}(E)]^n , \tag{18}$$

which converges for

$$D(E) > K , \tag{19}$$

where K^n is the number of self-avoiding walks of n steps. To correct for the neglect of the self-energy terms in the denominators, we replace K by z, the number of near neighbors.[5] This guarantees that for $v(R)$ set equal to 0, the band edges occur at $\pm z$, as they must in the tight-binding model.

As an example, let us apply Eq. (19) to the two-dimensional square lattice. The geometric mean of the energy denominator is given by

$$\begin{aligned}\ln D(E) &= \frac{1}{a^2}\int_{-a/2}^{a/2} dx \int_{-a/2}^{a/2} dy \ln\left| E - V_0 \cos\frac{2\pi}{a}x - V_0 \cos\frac{2\pi}{a}y \right| \\ &= \frac{1}{(4\pi)^2}\int_0^{2\pi} d\theta \int_0^{2\pi} d\varphi \ln\left| E - V_0\cos\varphi - V_0\cos\theta \right| \\ &= \frac{1}{\pi}\int_{\varphi_1}^{\pi} d\varphi \ln\left| \frac{|E - V_0\cos\varphi|}{2} + \tfrac{1}{2}[(E - V_0\cos\varphi)^2 - V_0^2]^{1/2} \right| + \frac{\varphi_1}{\pi}\ln\frac{V_0}{2} ,\end{aligned} \tag{20}$$

where

$$\cos\varphi_1 = (E - V_0)/V_0 \tag{21}$$

for $E \geq 0$. For $E = 0$, $D(E) = V_0/2$, and Eq. (19) shows that there are localized states for $V_0 > 2z$. As E moves away from the center of the band, $D(E)$ increases, making it easier to get localized states. Thus, from Eq. (20) we see that

$$V_0/2 < D(E) < 2V_0 . \tag{22}$$

This implies that for $z/2 < V_0 < 2z$ there exist mobility edges (that is, there are localized states nearer to the band edges and extended states nearer to the band center). It is not difficult to show that similar results occur in three dimensions.

In conclusion, the analysis presented here shows that in one dimension there is a transition from all states localized to most states extended, at a critical value of V_0. In two and three dimensions, there is a critical value of V_0 above which all states are localized. For smaller values of V_0, there exists a range of values of V_0 for which there are localized and extended states separated by mobility edges. For smaller values of V_0, all states are extended.

IV. APPLICATION TO AN EXPERIMENTAL SYSTEM

The present theory should be appropriate to metallic systems, with charge-density waves, in which the charge-density wave does not eliminate the entire Fermi surface. This is the case in the dichalcogenides, which are quasi-two-dimensional systems.[10] Most likely, however, the potential due to the charge-density waves is not strong enough to cause the predicted localization effects. As suggested by Azbel, the mercury chain compounds would be good candidates for looking for such effects.[1,11] In general, the effects discussed in this article should be observed in any metallic system containing a periodic potential incommensurate with the main lattice potential and comparable in strength to the width of the conduction band. The samples should be pure enough so that the effects are not masked by localization effects due to impurities.

A system to which the one-dimensional results should be applicable is a metallic or semiconducting system with a superlattice.[12] For example, consider the following tight-binding model for such a system:

$$\begin{aligned}\Sigma_{\vec{a}_\perp} f(\vec{R}_\perp + \vec{a}_\perp, z) + f(\vec{R}_\perp, z+b) + f(\vec{R}_\perp, z-b) \\ + v(z) f(\vec{R}_\perp, z) = E f(\vec{R}_\perp, z) ,\end{aligned} \tag{23}$$

where $\vec{R}_\perp$ labels positions of sites in atomic planes perpendicular to the modulation axis (the z axis), $\vec{a}_\perp$ labels near-neighbors in a plane, b gives positions of neighboring planes, and $v(z)$ is the periodic modulation potential. The Fourier transform in the $\vec{R}_\perp$ coordinate gives

$$\begin{aligned}g(\vec{k}, z+b) + g(\vec{k}, z-b) + v(z)\, g(\vec{k}, z) \\ = [E - \epsilon(\vec{k})]\, g(\vec{k}, z) ,\end{aligned} \tag{24}$$

where

$$g(\vec{k}, z) = \Sigma_{\vec{R}_\perp} e^{i\vec{k}\cdot\vec{R}_\perp} f(\vec{R}_\perp, z) ,$$

and

$$\epsilon(\vec{k}) = \Sigma_{\vec{a}_\perp} e^{i\vec{k}\cdot\vec{a}_\perp} .$$

From Eq. (1) and the previous section, we predict that for potential $v(z)$ stronger than a critical value there is localization perpendicular to the modulation, meaning that the system becomes a two-dimensional conductor. For weaker $v(z)$, there could be apparent mobility edges (since the modulation potential $v(z)$ has period much longer than a lattice constant) for conduction perpendicular to the modulation planes. Although in such superlattice systems the modulation potential is usually

commensurate with the lattice potential, if its period is many lattice constants long, it can be considered incommensurate for all purposes.

Another important application of the one-dimensional model is the application to an electron moving in a periodic lattice and in a magnetic field.[1] The fact that Eq. (1) has a continuous density of states in a regime in which the self-energy in k or r space has branch cut, however, does not mean that the discrete (and possibly "devil's-stair-type") spectrum predicted by Azbel for cyclotron resonance, de Haas–van Alphen effect, etc., does not occur. Azbel's effects are simply restricted to the region in which there are discrete states, (i.e., Landau levels).

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[1] M. Ya. Azbel, Phys. Rev. Lett. 43, 1954 (1979); Zh. Eksp. Teor. Fiz. 46, 929 (1963) [Sov. Phys.–JETP 19, 634 (1963)]; 46, 929 (1964) [19, 634 (1964)]; Dokl. Akad. Nauk. SSSR 159, 703 (1964) [Sov. Phys. Dokl. 5, 1549 (1964)].

[2] S. Aubry, in *Proc. Colloquium on Group Theoretical Methods in Physics*, March, 1979, Lectures in Physics (Springer, Berlin, in Press).

[3] J. B. Sokoloff, J. E. Sacco, and J. F. Weisz, Phys. Rev. Lett. 41, 1561 (1978).

[4] P. W. Anderson, Phys. Rev. 109, 1492 (1958).

[5] E. N. Economou and M. H. Cohen, Phys. Rev. B 5, 2931 (1972).

[6] E. N. Economou and M. H. Cohen, Phys. Rev. B 4, 396 (1972).

[7] K. M. Watson, Phys. Rev. 105, 1388 (1957).

[8] H. S. Wall, *Analytic Theory of Continued Fractions* (Chelsea, New York, 1948), pp. 14–16.

[9] A. Ya. Khintchine, *Continued Fractions* (Noordhoff, Groningon, 1963), pp. 10—11.

[10] J. A. Wilson, F. J. DiSalvo, and S. Mahajan, Adv. Phys. 24, 117 (1975).

[11] C. K. Chiang, R. Spal, A. Denenstein, A. J. Heeger, N. D. Miro, and A. G. MacDiarmid, Solid State Commun. 22, 293 (1977).

[12] R. Dingle, H. L. Storma, A. C. Gossand, and W. Wieymann, Inst. Phys. Conf. Ser. No. 45, Ch. 3, p. 248, 1978 (and references contained therein).