Mobility Gap and Anomalous Dispersion*

Daniel C. Mattis

and

Fumiko Yonezawa†

Belfer Graduate School of Science, Yeshiva University, New York, New York 10033

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It is shown that anomalous dispersion of quasiparticles leads to nonpropagating states. Therefore regions of anomalous dispersion define a sort of "mobility gap." Using the coherent-potential approximation, we calculate the conditions for obtaining such a mobility gap in a disordered binary alloy. Just outside the mobility edges located at ω_c the mobility vanishes as $|\omega-\omega_c|$.

We consider the question of the existence of a "mobility gap," of whether in a disordered medium there is a range of allowed energy levels which are all nonpropagating states. This concept, first conjectured by Mott,[1] Cohen, Fritzsche, and Ovshinsky,[2] and others, is of great physical interest in the electronic band structure of liquids, amorphous semiconductors, and disordered alloys. It has been shown that in the disordered linear chain *all* states are localized,[3] so that the "mobility gap" could well include all allowed energy levels. In the case of the three-dimensional disordered medium one needs a specific criterion for the existence and location of such a gap. Criteria based on probabilistic concepts have been put forward.[4]

It is the purpose of the present work to present a different sort of criterion, based on the dispersion of quasiparticles. We shall show that if there is a region of *anomalous* dispersion, ordinary wave packets made with states in this region cannot propagate, and we shall therefore identify this region of energy states as constituting a mobility gap. We shall then use the well-known coherent-potential approximation (CPA)[5] to obtain the quasiparticle energies in a special case, as an example to demonstrate conditions under which one finds anomalous dispersion.

Consider a wave packet about an average energy ω_0:

$$F(\vec{r}, t) \equiv \int_{-\infty}^{\infty} d^3k\, e^{i\vec{k}\cdot\vec{r}} \int_{-\infty}^{\infty} d\omega\, f(\omega - \omega_0) e^{-i\omega t}/[\omega - \zeta_k(\omega) - \epsilon_k], \tag{1}$$

where $f(\omega - \omega_0)$ is a given envelope function. With the initial condition that it be centered near the origin at $t=0$, the wave packet propagates out as time goes forward. This causality requirement determines the sign of $\Gamma_k(\omega)$, the imaginary part of the self-energy $\zeta_k(\omega)$:

$$\zeta_k(\omega) \equiv R_k(\omega) - i\Gamma_k(\omega), \quad \Gamma_k(\omega) \geq 0. \tag{2}$$

If Γ is not too large, the above integrand is approximated by a pole located at $\omega_k - i\Gamma_k(\omega_k)$, where ω_k is defined as the solution of

$$\omega_k - R_k(\omega_k) - \epsilon_k = 0. \tag{3}$$

Now assuming $f(\omega - \omega_0)$ to be narrowly peaked about $\omega = \omega_0$, we may expand about ω_0 as follows:

$$\begin{aligned} \omega - \zeta_k(\omega) &= \omega_0 - R_k(\omega_0) + i\Gamma_k(\omega_0) + (\omega - \omega_0)[1 - \partial R_k(\omega_0)/\partial\omega_0 + i\partial\Gamma_k(\omega_0)/\partial\omega_0] + \cdots \\ &= \epsilon_{k_0} + i\Gamma_0 + (\omega - \omega_0)\mu^* + \cdots, \end{aligned} \tag{4}$$

where $\epsilon_{k_0} \equiv \omega_0 - R_k(\omega_0)$, $\mu^* \equiv 1 - \partial R_k(\omega_0)/\partial\omega_0$, and the small correction terms $O(i(\omega - \omega_0)\,\partial\Gamma/\partial\omega)$ (it can be assumed that Γ is slowly varying) and $O((\omega - \omega_0)^2)$ are neglected.

We now average over a large sphere at r:

$$F(r, t) = \exp(-i\omega_0 t)\int d\epsilon_k\, \rho_0(\epsilon_k)\, \frac{\sin kr}{kr} \int d\Omega\, \exp(-i\Omega t)\, \frac{f(\Omega)}{(\epsilon_{k_0} - \epsilon_k) + \Omega\mu^* + i\Gamma_0}, \tag{5}$$

where $\Omega \equiv \omega - \omega_0$. We further expand k about k_0, suitably averaging over angles if necessary,

$$k = k_0 + (\partial\epsilon/\partial k)_0^{-1}(\epsilon_k - \epsilon_{k_0}) = k_0 + z/V_k, \tag{6}$$

which serves to define $V_0 \equiv (\partial\epsilon/\partial k)_0$ and $z \equiv \epsilon_k - \epsilon_{k_0}$. Taking slowly varying factors outside the k integral, we have

$$F(r, t) = \exp(-i\omega_0 t)\rho_0(\epsilon_{k_0})(2ik_0 r)^{-1}\int d\Omega\, f(\Omega)\exp(-\Omega t)[I_1(\Omega) - I_2(\Omega)], \tag{7}$$

where the integrals

$$\begin{aligned} I_1(\Omega) &= \exp(ik_0 r)\int dz\, \exp(izr/V_0)(\Omega\mu^* + i\Gamma_0 - z)^{-1}, \\ I_2(\Omega) &= \exp(-ik_0 r)\int dz\, \exp(-izr/V_0)(\Omega\mu^* + i\Gamma_0 - z)^{-1} \end{aligned} \tag{8}$$

can be evaluated by contour integration. For electronlike particles, $V_0 > 0$ and the contour for I_1 is closed by an infinite semicircle in the upper-half complex z plane and I_2 in the lower half. For holelike particles, $V_0 < 0$ and the respective contours are interchanged. Thus,

$$F(r, t) = -\exp(-i\omega_0 t \mp ik_0 r)[\pi\rho_0(\epsilon_{k_0})/k_0 r]\tilde{f}(t - \mu^* r/|V_0|)\exp(-r\Gamma_0/|V_0|), \tag{9}$$

in which $\tilde{f}(\tau) \equiv \int d\Omega\, f(\Omega)\exp(-i\Omega\tau)$. Except for the $\mp$ sign in the phase factor, this result is independent of the sign of V_0.

The sign of the dispersion parameter μ^* is, however, crucial. For $\mu^* > 0$, $F(r, t)$ is an outgoing spherical wave packet, decaying exponentially as it leaves the origin, because of the incoherent scattering. For $\mu^* < 0$, it becomes an *incoming* spherical wave packet, which *grows* unphysically as it approaches the origin and then ceases to exist at large t. The conclusion that there is a sink at $r=0$, i.e., that the eigenstates are everywhere localized, becomes inescapable. This is in accord with the usual understanding of anomalous dispersion in optics, a familiar phenomenon observed in a narrow range of frequencies near resonant absorption.

It is instructive to consider the example of a retarded Green's function,[6]

$$G_{\rm ret}(\vec{r},t) = -i\theta(t)\langle\{\psi(r,t),\ \psi^\dagger(0,0)\}\rangle = \int_{-\infty}^{\infty} d\omega\, e^{-i\omega t} G(\omega + i0^+), \tag{10}$$

in which $\theta(t)=1$ for $t>0$ and vanishes at $t<0$, and

$$G(\omega) = \int d^3k \int d^3k'\, e^{i\vec{k}\cdot\vec{r}} \langle\langle C_k | C_{k'}{}^\dagger\rangle\rangle_\omega. \tag{11}$$

We must average over the ensemble of random configurations in the disordered medium, thus restoring translational invariance of a sort:

$$\bar{G}(\omega) = (2\pi)^{-1}\int d^3k\, e^{i\vec{k}\cdot\vec{r}}[\omega - \zeta_k(\omega) - \epsilon_k]^{-1}, \tag{12}$$

where the self-energy function $\zeta_k(\omega)$ is real for real ω, and is complex, $\zeta(\omega \pm i0^+) = R \mp i\Gamma$, with $\Gamma > 0$, for ω just off the real axis. The branch cut is well approximated by a simple pole, as in Eq. (3). Our previous discussion makes it clear that regions of anomalous dispersion will not contribute significantly to $G_{\rm ret}(\vec{r},t)$. A spectral resolution of $G(\vec{r},t)$ at large r will not show any component belonging to frequencies within this region, corresponding to the notion of a "mobility gap."

It remains to calculate $R_k(\omega)$ and $\Gamma_k(\omega)$, and the CPA now provides a convenient and fairly accurate method for doing so. We take a specific model: the random binary alloy with potential energy $\pm\frac{1}{2}U$ at each site. The CPA equations are

$$\langle\langle C_k | C_{k'}{}^\dagger\rangle\rangle_\omega = (2\pi)^{-1}\delta_{k,k'}[\omega - \zeta(\omega) - \epsilon_k]^{-1}, \tag{13}$$

where the average T matrix is made to vanish:

$$\bar{T}(\omega) = \frac{\frac{1}{2}U - \zeta(\omega)}{1 - [\frac{1}{2}U - \zeta(\omega)]g(\omega)} + \frac{-\frac{1}{2}U - \zeta(\omega)}{1 - [-\frac{1}{2}U - \zeta(\omega)]g(\omega)} = 0, \tag{14}$$

where

$$g(\omega) = N^{-1}\sum_k[\omega - \zeta(\omega) - \epsilon_k]^{-1} = \int d\epsilon\, \rho_0(\epsilon)[\omega - \zeta(\omega) - \epsilon]^{-1}. \tag{15}$$

These yield a self-energy function $\zeta(\omega) = R(\omega) - i\Gamma(\omega)$ (independent of k in the CPA). Supposing the unperturbed bandwidth to be Δ, we introduce dimensionless units such that $U=1$ and $\delta = \Delta/U$, and approximate ρ_0 by the semicircular function,

$$\rho_0(\epsilon) = (4/\pi\delta)[1 - (2\epsilon/\delta)^2]^{1/2}, \quad |\epsilon| < \tfrac{1}{2}\delta, \tag{16}$$

for which a simple algebraic expression can be obtained for $g(\omega)$:

$$g(\omega) = 8\delta^{-2}\{\omega - \zeta - [(\omega-\zeta)^2 - \tfrac{1}{4}\delta^2]^{1/2}\}. \tag{17}$$

Combining the above with Eq. (14) we obtain a cu-

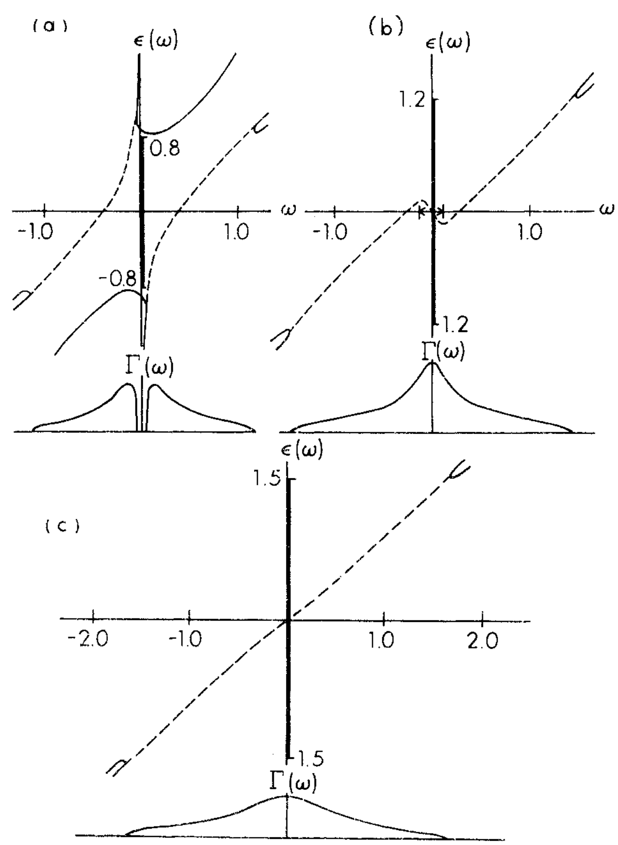

FIG. 1. Dispersion relations, $\epsilon(\omega)$ versus ω, for (a) $\sigma=1.6$, (b) $\sigma=2.4$, and (c) $\sigma=3.0$. Solid lines, real solutions of the cubic equation; dashed lines, real parts of the complex conjugate solutions for ζ; thick line on the $\epsilon(\omega)$ axis, allowed region over which the variable ϵ_k is defined. At the bottom of each figure is shown the imaginary part $\Gamma(\omega)$ of ζ. (a) corresponds to a separated-band case with a finite density-of-states gap, while (b) and (c) are examples with no density-of-states gaps. (b) shows anamalous dispersion, with a mobility gap indicated by an arrow.

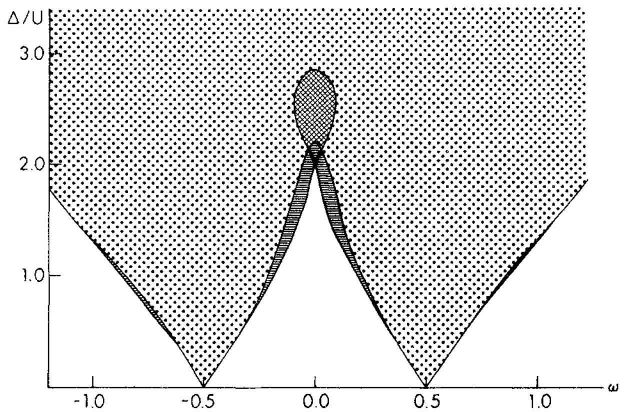

FIG. 2. Dependence of the quasiparticle spectrum and and the mobility gaps upon $\sigma = \Delta/U$ and ω. The density of states is nonvanishing in the dotted regions. Regions of localized states within our mobility gap are cross hatched, and those within the Economou-Cohen gap are single hatched.

bic equation in ζ:

$$\omega\zeta^3 - \tfrac{1}{4}(1 - \tfrac{1}{4}\delta^2)\zeta^2 - \tfrac{1}{4}\omega\zeta + \tfrac{1}{16} = 0. \tag{18}$$

This is solved numerically to yield $\epsilon(\omega) \equiv \omega - R(\omega)$. When $\epsilon(\omega) = \epsilon_k$, we have located the pole. This is plotted in Fig. 1. For $\delta^2 < 4$, a density-of-states gap opens up about $\omega = 0$ and there is no region of anomalous dispersion. In the range $4 < \delta^2$ one can expand Eq. (17) in powers of ω, and obtain an expression for $\partial\epsilon(\omega)/\partial\omega$ at $\omega = 0$. This quantity turns out negative only in the narrow range $4 < \delta^2 < 8$, and anomalous dispersion again ceases to exist at $\delta^2 > 8$.

It follows from the preceding arguments that the states indicated by an arrowed portion on the ω axis in Fig. 1(b) are localized and that the mobility edges at ω_c are determined by the vanishing of $d\epsilon(\omega)/d\omega$. The mobility gap obtained in this manner is shown in Fig. 2 by cross hatching. In Fig. 2 we contrast our computed regions of anomalous dispersion to the regions of localized states predicted by the probabilistic criterion of Economou and Cohen. The two criteria agree only near $\delta = 2$. This discrepancy appears to be worthy of further study, for it suggests that there may be more than one way to localize waves within a continuum.

Our formulation gives us an insight into the behavior of the mobility edges. Although the mobility is not identically zero for $|\omega| > \omega_c$, it approaches zero when $|\omega| \to |\omega_c|$ because the dispersion parameter $\mu^* = [d\epsilon(\omega)/d\omega]^{-1}$ increases and goes to infinity at the critical energy. As we have $\epsilon(\omega) - \epsilon(\omega_c) \propto (\omega - \omega_c)^2$ near $|\omega_2|$, $1/\mu^* \propto |\omega - \omega_c|$. The mobility μ is defined by $\mu = e^2\tau/\mu^*$. The lifetime τ of a quasiparticle is inversely proportional to $\Gamma(\omega)$, which can now be replaced by $\Gamma(\omega_c)$ since deviation from this value only gives a higher-order correction. As a result, the mobility near the critical point is seen to vanish linearly as $\mu \propto |\omega - \omega_c|$.

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†On leave from the Department of Applied Physics, Tokyo Institute of Technology, Meguroku, Tokyo 152, Japan.

[1]N. F. Mott, Advan. Phys. 16, 49 (1967).

[2]M. H. Cohen, H. Fritzsche, and S. Ovshinsky, Phys. Rev. Lett. 22, 1065 (1969).

[3]R. Makinson and A. Roberts. Proc. Phys. Soc., London 79, 222 (1962); R. Borland, Proc. Roy. Soc., Ser. A 274, 529 (1963).

[4]E. N. Economou and M. H. Cohen, Phys. Rev. B 5, 293 (1972).

[5]P. Soven, Phys. Rev. 156, 809 (1967); F. Yonezawa, Progr. Theor. Phys. 40, 734 (1968); B. Velicky, S. Kirkpatrick, and H. Ehrenreich, Phys. Rev. 175, 747 (1968).

[6]D. N. Zubarev, Usp. Fiz. Nauk 71, 71 (1960) [Sov. Phys. Usp. 3, 320 (1960)]. The symbols used in these equations are defined by Zubarev.