

## Letters to the Editor

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### On the Interaction of Conduction Electrons with Lattice Vibrations

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BARDEEN<sup>1-4</sup> and Fröhlich<sup>5,6</sup> have recently proposed theories of superconductivity based on interactions between conduction electrons and the zero-point vibrations of the lattice. In his development, Bardeen views this interaction as giving rise to Brillouin zone boundaries with small energy gaps ( $\sim kT_c$ ) at the Fermi surface of the electron distribution. Electrons with energies near the Fermi surface have small effective mass in consequence of the high curvature in their energy *versus* wave number relation at a zone boundary. Bardeen suggests that these are the superconducting electrons and indicates that it is possible to give a consistent interpretation for properties of superconductors in terms of the large diamagnetism exhibited by a gas of non-interacting electrons with small effective mass.

In this note we study the effective mass of conduction electrons which interact with the zero-point lattice vibrations. Fröhlich has calculated, with second-order perturbation theory, the self-energy of an electron interacting with the lattice normal modes of vibration (the phonon field). The energy denominators vanish and this perturbation procedure becomes invalid for virtual intermediate states which conserve energy.<sup>7</sup> Bardeen has avoided this difficulty with a variational treatment that leads to band structure in the energy *versus* wave number curve for the electrons. However, this result is obtained only after a questionable approximation for the mean energy of the virtual intermediate electron plus phonon states. We have calculated the energy of electrons interacting with the lattice vibrations by means of a Bloch-Nordsieck-type canonical transformation.<sup>8</sup> The B-N treatment is particularly suited to this type of problem, in which the energies associated with the crystal normal modes that are excited at low temperatures are very small compared with the electron energies. In this case neglect of electron recoil upon scattering with phonons introduces but a small error. Perturbation theory is avoided and, along with it, the vanishing energy denominators. We interpret our result as arguing against the conclusion that electrons have small effective mass in consequence of their interactions with the lattice normal modes.

The hamiltonian for electrons moving in a periodic lattice potential and interacting with normal modes of the crystal may be written as in reference 1. Neglect of the electrons' recoil upon scattering with the phonon field gives a hamiltonian that is linear in the electron momentum operators  $-i\hbar \text{grad}_r$ . The Schrödinger equation with this hamiltonian is exactly soluble by means of a canonical transformation of the type introduced originally by Bloch and Nordsieck<sup>9</sup> in their study of the "infrared catastrophe," and used more recently in attempts at nonperturbation solutions for meson problems.<sup>9</sup> The eigensolutions correspond to a product of Bloch functions for individual electrons moving in a periodic lattice potential, multiplied by hermite functions for displaced zero-point oscillations of the phonon field, and multiplied by a factor expressing the interactions between the different electrons in consequence of their coupling with the lattice vibrations. The energy eigenvalues differ from those of the band theory for elec-

trons moving in the periodic potential of the static lattice in the following way. In the periodic potential of a static lattice with period  $\mathbf{a}$ , there occur forbidden energy bands for electrons with momenta in the neighborhood of  $m\mathbf{v}_i = \pi\hbar(\mathbf{n}_i/\mathbf{a}_i)$ , where the  $\mathbf{n}_i$  are integers. Electrons with these momenta suffer Bragg reflection. The lattice vibrations have the period  $\mathbf{L}$ , the crystal dimensions, and they superimpose, on the above bands, forbidden energy bands for electrons which satisfy an analogous Bragg condition with the lattice vibrations,

$$\omega_f^2 = [\mathbf{v} \cdot (\mathbf{\Lambda}_f - \mathbf{k}_n)]^2. \quad (1)$$

Here  $\omega_f$ , the angular frequency of the lattice vibrations with wave vector  $\mathbf{\Lambda}_f = 2\pi(f_1/L_1, f_2/L_2, f_3/L_3)$ , is given by  $\omega_f = s\Delta_f$ ;  $\mathbf{s}$  is the velocity of sound in the crystal, the  $f_i$  are integers, and  $\mathbf{k}_n = 2\pi(n_1/a_1, n_2/a_2, n_3/a_3)$  is a reciprocal lattice vector.<sup>10</sup> The width of these forbidden bands is

$$\sim (\text{amplitude of the lattice potential}) / (\text{number of unit cells in the crystal})^{\frac{1}{2}}.$$

It is thus very small. These narrow forbidden bands occur throughout the electron distribution at momentum values that satisfy the condition in Eq. (1). They do not critically change the energy levels of the conduction electrons and hence do not critically alter their effective mass. A first-order perturbation treatment of the neglected electron recoil yields a small correction ( $\sim s/v$ ) to these results.

In summary, our calculations do not indicate a small effective mass for electrons in consequence of their interactions with lattice vibrations. The energy shell distribution as proposed by Fröhlich does not appear to be energetically preferred. When other important factors such as the interelectron coulomb repulsion are considered, one may hope to find an adequate theory of superconductivity developed along lines of these very attractive physical proposals of Bardeen and Fröhlich. A manuscript discussing the details of this calculation is in preparation. Valuable discussions with Professor L. I. Schiff are gratefully acknowledged.

<sup>1</sup> J. Bardeen, Phys. Rev. **80**, 567 (1950).

<sup>2</sup> J. Bardeen, Phys. Rev. **81**, 469 (1951).

<sup>3</sup> J. Bardeen, Phys. Rev. **81**, 829 (1951).

<sup>4</sup> J. Bardeen, Phys. Rev. **81**, 1070 (1951).

<sup>5</sup> H. Fröhlich, Phys. Rev. **79**, 845 (1950).

<sup>6</sup> H. Fröhlich, Proc. Phys. Soc. (London) **A64**, 129 (1951).

<sup>7</sup> Fröhlich takes principal values in the integration over intermediate states.

<sup>8</sup> F. Bloch and A. Nordsieck, Phys. Rev. **52**, 54 (1937).

<sup>9</sup> Lewis, Oppenheimer, and Wouthuysen, Phys. Rev. **73**, 127 (1948); S. D. Drell, Phys. Rev. **83**, 555 (1951).

<sup>10</sup> These bands do not appear in Fröhlich's work because he takes principal values for his integrals over the intermediate states.

### Anomalous Intensity Distribution of Rotation Lines in Fundamental Vibration Bands in Triatomic Molecules\*

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A MECHANISM will be discussed here to account for the anomalous intensity distribution of the rotation lines in certain fundamental bands in the infrared spectra of triatomic molecules, notably in the 3.8- $\mu$  band in the H<sub>2</sub>S spectrum. The theory assumes that it is necessary to take into account the mixing of the wave functions for a given rotation state in a vibration level with the wave functions of rotation states in other vibration levels in order to calculate the intensities with accuracy. The mixing must be between wave functions of the first excited states of the three vibration frequencies to give a first-order effect. Such a mixing may come about through the coupling of the vibrations through the Coriolis operator  $p_z P_z / I_{zz}$ , the  $z$  axis having been chosen as the axis of spin of the molecule. The mixed wave function