

Quantization and Stability of Currents in Superconductors

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In an electron gas with pairing correlations there exists the possibility of excited states corresponding to a modified pairing of the electrons and associated with "supercurrents." The quantization of these states, which is also reflected in the quantization of flux through a multiply connected superconductor, implies the occurrence of macroscopic "isomerism" providing an immediate explanation of the persistent currents. It is suggested that similar phenomena, associated with vortex lines in the electron fluid, may occur in simply connected samples under suitable conditions.

THE recent experimental discovery¹ that the flux passing through a hole in a superconducting body is quantized in units of $hc/2e$ brings into focus the peculiar phenomenon of quantization of supercurrents with macroscopic values of the angular momentum. The quantization of the flux had been predicted² and has recently been further discussed.^{3,4} In this article we shall consider its relation to the energy spectrum of the superconductor and its role in ensuring the stability of the persistent currents.

The modes of excitation of a superconductor have been extensively discussed in the recent literature.⁵ One has especially considered, in addition to the lattice vibrations, the excitation of quasi-particles and certain collective modes, such as plasmons (density fluctuations) and excitons (bound pairs of quasi-particles with finite relative angular momentum).

In addition, there may be quantum states with a pairing of the electrons different from the one which is normally assumed and which involves time-reversed single-electron states. If in the new pairing the two particles have a different density distribution, the pairing energy will in general be reduced by a finite amount; for example, if in a cubic box one pairs the states $n_x\uparrow$ with $(n_x+1)\downarrow$, the pairing matrix element for a short-range attraction is reduced by a factor of 2. The resulting states of the metal will therefore have such high energy that they may be expected to damp quickly into other modes with a more efficient pairing.

The pairing correlation will be preserved if all the single-particle wave functions are multiplied by a phase factor corresponding to a total wave function

$$\Psi = \exp\{i \sum_j \chi(\mathbf{r}_j)\} \Psi_0, \quad (1)$$

where Ψ_0 represents the ground state.

In discussing the properties of the states of type (1) we shall first neglect all magnetic effects. These effects, although of major importance for a great many properties of the superconductor, do not affect the essential structure of the quantum states and therefore may be simply added later.

In the absence of magnetic coupling the current and energy in the states (1) are given by

$$\mathbf{i}(\mathbf{r}) = (e\hbar/\mu)n(\mathbf{r})\nabla\chi(\mathbf{r}) \quad (2)$$

and

$$E = E_0 + \frac{\hbar^2}{2\mu} \int n(\mathbf{r})(\nabla\chi)^2 d\tau, \quad (3)$$

where $n(\mathbf{r})$ is the density of electrons and where E_0 is the energy of the ground state, which has been assumed to have a vanishing current density. Thus, the states (1) correspond to a collective flow described by the velocity potential $(\hbar/\mu)\chi(\mathbf{r})$. For (1) to represent a stationary state it is a necessary condition that

$$\nabla \cdot \mathbf{i} = 0 = \nabla \cdot (n\nabla\chi). \quad (4)$$

If χ is single valued, as it must be for a simply connected piece of metal, we have from (4),

$$\int n(\nabla\chi)^2 d\tau = \frac{\mu}{e\hbar} \int \chi \mathbf{i} \cdot d\mathbf{f}, \quad (5)$$

where $d\mathbf{f}$ is a surface element of the conductor. Thus, if a current is not flowing in and out of the metal, we only have the trivial solution $\chi = \text{const}$.

We can therefore obtain solutions of the type (1) only by either having currents flowing through the ends of a superconducting piece of metal or by having a multiply connected geometry.

MULTIPLY CONNECTED SUPERCONDUCTOR

The solution to (4) is characterized by the total change of χ on circling the tunnels in the supercon-

¹ B. S. Deaver, Jr. and William M. Fairbank, *Phys. Rev. Letters* **7**, 43 (1961); R. Doll and M. N abauer, *Phys. Rev. Letters* **7**, 51 (1961).

² F. London, *Superfluids* (John Wiley & Sons, Inc., New York, 1950), p. 152.

³ N. Byers and C. N. Yang, *Phys. Rev. Letters* **7**, 46 (1961).

⁴ L. Onsager, *Phys. Rev. Letters* **7**, 50 (1961).

⁵ For a review, see J. Bardeen and J. R. Schrieffer, *Progress in*

Low-Temperature Physics, edited by J. C. Gorter (North Holland Publishing Company, Amsterdam, 1961), Vol. III, p. 170.

ductor, and the requirement that the wave function (1) be univalued implies that the states are quantized with the increments in χ being equal to $2\pi\nu$ with ν an integer.

As a prototype, we consider a hollow cylinder (inside radius R_1 outside radius $R_2=R_1+d$, and length L) and also assume that $n(\mathbf{r})$ for the conduction electrons is a constant inside the metal, corresponding to a uniform smearing out of the positive ions. The solutions to (4) with the required boundary conditions are then simply

$$\chi(\mathbf{r}) = \nu\varphi; \quad \nu = 0, \pm 1, \pm 2, \dots \quad (6)$$

The states (1) thus correspond to a modified pairing, with the electrons occupying the states

$$\sin\left(\frac{\pi n_z z}{L}\right) j_{m,n_r}(r) \times \begin{cases} e^{i(m+\nu)\varphi}\alpha, \\ e^{-i(m-\nu)\varphi}\beta, \end{cases} \quad (7)$$

with α and β representing the spin functions and $j_{m,n_r}(r)$ the radial wave functions satisfying

$$\left(-\frac{\hbar^2}{2\mu} \frac{1}{r} \frac{\partial}{\partial r} r \frac{\partial}{\partial r} + \frac{\hbar^2 m^2}{2\mu r^2}\right) j_{m,n_r}(r) = \epsilon_{m,n_r} j_{m,n_r}(r). \quad (8)$$

Each pair has angular momentum $2\hbar\nu$.

It is also possible to have a pairing with odd-integral amounts of angular momentum per pair. Such pairings correspond to occupation of the states $(m+\nu+\nu')\uparrow$ and $-(m-\nu+\nu')\downarrow$, and the total wave function can be written

$$\Psi_{\nu,\nu'} = \exp\{i \sum_j (\nu + \sigma_{z_j} \nu') \varphi_j\} \Psi_0. \quad (9)$$

The condition that Ψ be univalued now implies that $\nu \pm \nu'$ be integers, or that ν and ν' either be both integers or both half-integers.

One may somewhat improve the wave functions (9) by allowing the radial wave function j_{m,n_r} to depend on ν and ν' . As long as the two members of the pair have the same radial functions, the pairing energy is not affected, but the sum of the kinetic energies in the radial motion for the two electrons has a minimum if the radial function satisfies the Eq. (8) with m^2 replaced by $(m+\nu')^2 + \nu^2$ which, to within higher order effects, is equivalent to the replacement $m \rightarrow m + \nu'$. For such a wave function, the total energy is, to leading order in N , independent of ν' , and is given by

$$E_\nu = (\hbar^2/2\mu) \langle 1/r^2 \rangle N \nu^2, \quad (10)$$

where N is the total number of conduction electrons.

With this readjustment of the radial wave functions, states having the same ν and differing only by integral amounts in ν' , become identical, since they correspond to a pairing in the same set of single-particle states. The spectrum of quantum states with supercurrents is thus given by

$$\Psi_\nu = \exp\{i\nu \sum_j \varphi_j\} \times \begin{cases} \Psi_0, & \nu = 0, \pm 1, \dots, \\ \Psi'_0, & \nu = \pm \frac{1}{2}, \pm \frac{3}{2}, \dots, \end{cases} \quad (11)$$

where Ψ'_0 is a wave function of the same structure as Ψ_0 , but for which the single-particle states have half-integral, rather than integral values of m .

The total angular momentum along the axis for the states (11) is, in units of \hbar ,

$$M = \nu N. \quad (12)$$

The energy per unit of angular momentum is very small. Thus, in quasi-particle excitations, the minimum energy per angular momentum is of the order

$$\Delta/m \gtrsim \Delta/k_F R_2, \quad (13)$$

where Δ is the energy gap and $\hbar k_F$ the Fermi momentum, and thus exceeds that corresponding to (10) by a factor

$$k_F d (\Delta/E_F) \sim d/\xi_0, \quad (14)$$

where $\xi_0 [\sim (\hbar^2 k_F / \mu \Delta)]$ is the coherence distance for the pairs. For very small dimensions ($d \lesssim \xi_0$), the relevant quasiparticle excitation energies become greater than Δ , and the factor corresponding to (14) remains greater than unity.

The energy of the lowest quantum state of the superconductor with given angular momentum thus exhibits a behavior illustrated in Fig. 1. The existence of the states of the type (11) implies the occurrence of *macroscopic isomerism*.

These isomeric states, associated with supercurrents, may be expected to have a remarkable stability, since a change in the quantum number ν involves a macroscopic modification of the system. The highest angular momentum involved in a single electron transition, or plasmon excitation, is $\sim k_F R$, and hence a change in ν involves simultaneous excitations to order $(k_F d) \cdot (k_F L)$. Thus, the usual relaxation mechanisms would seem to give effects proportional to some perturbation parameter to this high order; it also seems practically impossible to construct external perturbations which would change the quantum number ν .

Thus, the quantization of the supercurrent with macroscopic values of the angular momentum seems to offer an immediate explanation of the dramatic phenomenon of the persistent currents in multiply connected superconductors. It seems appropriate to think of the states with different ν as representing different phases of the superconducting system.

It is important to emphasize that the stability of these states depends essentially on the presence of the pairing correlations, which drastically suppress all excitation modes except the special ones (11) in which the densities remain unchanged. In the absence of pairing correlations, it becomes favorable to adjust the wave functions (11) so that the individual single-particle functions become stationary one-particle states. The total wave function then represent a definite configuration of single-particle excitations which can each decay independently, leading continuously to the

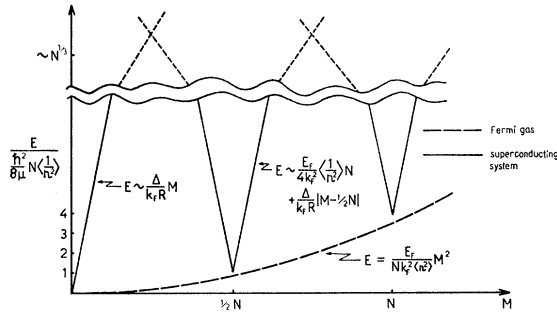


FIG. 1. Energy required to excite states of high angular momentum in a superconducting tube. The figure schematically illustrates the excitation energy of the lowest state of angular momentum M . The full-drawn curve refers to the spectrum of the superconductor, while the dashed curve shows the behavior of a normal Fermi gas. We have assumed dimensions such that $R \sim d \sim L$ and, thus, $k_F R \sim N^{1/3}$. The slopes of the full-drawn and dashed curves therefore differ by many orders of magnitude for a macroscopic system. The curves should exhibit a fine structure for angular momentum intervals $\sim N^{1/3}$ corresponding to the structure of the single-particle (and quasi-particle) spectra, but such a fine structure is not reproducible on the scale of the figure.

The curve for the normal metal can be described in terms of an effective moment of inertia equal to the rigid-body value. The superfluid can rotate with the same ease only for the special values $M = \nu N$. Actually, the moment of inertia for these states is somewhat less than the rigid-body value, as a result of the difference between irrotational and rigid flow. This difference is proportional to the difference between $\langle r^{-2} \rangle$ and $\langle r^2 \rangle^{-1}$ and vanishes in the limit of a thin tube ($d \ll R$).

The occurrence in the superconductor of the isolated low lying states with $M = \nu N$ implies the existence of isomeric states with practically unlimited stability. The characterization of these isomers in terms of the angular momentum refers to the special case of axial symmetry; more generally, any multiply connected domain will have isomers characterized by integer values of the circulation around the tunnels.

The spectrum is drawn without inclusion of the magnetic interaction effects. As discussed in the text, this interaction does not appreciably modify the structure of the wave functions, but the attraction between the current loops implies a further stabilization of the isomeric states. The energy with inclusion of this effect [see estimate (23)] corresponds, on the scale of the figure, to values of the order of the square of the ratio of the London penetration depth λ_L to the dimensions of the system.

ground state. In the case of an insulator, the readjusted wave function simply equals Ψ_0 .

For finite temperatures, the distribution of quasi-particles depends on the quantum number ν , since the displacement of the Fermi sphere favors quasi-particles moving in the opposite direction. For $\Delta = 0$, this "back flow" cancels the supercurrent, reflecting the instability of the state with displaced Fermi surface. For $\Delta \neq 0$, the back flow represents only a fraction of the supercurrent.

An interesting question arises if one considers tubes with smaller and smaller inner radius. Finally, the centrifugal energy per particle in the central region becomes comparable with the energy difference between the superconducting and normal phase, with the result that the superconductivity is destroyed inside a region extending to a radius $r_c \sim \nu \xi_0$. The energy per unit angular momentum in such a vortex line is still very much less than that associated with quasi-particle

excitations. Thus, the interesting possibility arises of having such free vortex lines with associated persistent currents in a simply connected superconductor. The estimate of r_c is somewhat modified by the magnetic coupling effects, which are also important in a discussion of the conditions under which such vortex lines may be observed (see below).

The states with quantized supercurrents are in many respects analogous to the states of quantized potential flow⁶ in liquid He II. Recently, experimental evidence⁷ has been obtained for the quantization of the circulation $\oint \mathbf{v} \cdot d\mathbf{s}$ in multiply connected domains in units of h/μ_{He} ; the corresponding states also appear to have essentially unlimited stability. In addition, it appears that quantized free vortex lines may occur in He II.⁷

CURRENTS THROUGH SIMPLY CONNECTED SUPERCONDUCTORS

If electrons flow in at one end and out at the other end of the superconductor the boundary conditions for χ are modified. For a simply connected superconductor the solution to (4) is again unique. As a prototype we consider a straight wire and neglect the variation in $n(\mathbf{r})$; the velocity potential is then given by

$$\chi(\mathbf{r}) = kz, \quad (15)$$

where $(e\hbar/\mu)kn$ is the current flowing through the wire. The state (1) now corresponds to a pairing of the electrons with each pair carrying a momentum $2\hbar k$.

The situation is here essentially different from that of the supercurrents flowing in a multiply connected domain, since χ is uniquely determined by the boundary conditions which can be varied continuously. There is thus no analog to the quantized isomeric states with the persistent currents. The suppression by the pairing correlation of all degrees of freedom, except that corresponding to uniform flow, implies that the usual relaxation mechanisms are almost ineffective. Still, the permissibility of infinitesimal changes in k leaves open the possibility of damping mechanisms which are much more effective than for the case of the persistent ring currents.

MAGNETIC EFFECTS

We first discuss the problem of the electron motion in the presence of a prescribed magnetic field and later return to the question of the self-consistency between this field and the currents it induces.

Thus, we consider a hollow superconducting cylinder with a given flux Φ passing through it; the magnetic field is assumed to vanish inside the conductor.

⁶ L. Onsager, Suppl. Nuovo cimento **6**, 249 (1949); R. P. Feynman, *Progress in Low-Temperature Physics*, edited by J. C. Gorter (North Holland Publishing Company, Amsterdam, 1955), Vol. I, p. 17.

⁷ W. F. Vinen, *Progress in Low-Temperature Physics*, edited by J. C. Gorter (North Holland Publishing Company, Amsterdam, 1961), Vol. III, p. 1.

If the flux is turned on adiabatically the wave functions (11) remain unaltered, but the energy and the total current density become

$$E_\nu = (\hbar^2/2\mu)\langle 1/r^2 \rangle N(\nu - \sigma)^2 \quad (16)$$

and

$$i(r) = (e\hbar/\mu)n(1/r)(\nu - \sigma), \quad (17)$$

where

$$\Phi = (hc/e)\sigma. \quad (18)$$

Although the stationary single-particle wave functions would be slightly modified by the presence of the flux,^{3,8} the pairing interaction suppresses such an adjustment, since the loss in pairing energy would far exceed the small gain in single-particle energies.⁹ The optimum radial wave functions are the solution of (8) with $m \rightarrow [(m + \nu')^2 + (\nu - \sigma)^2]^{1/2} \approx m + \nu'$.¹⁰

The expression (17) for the current shows that the self-consistent values of the flux, which lead to a vanishing current inside the superconductor, are given by $\sigma = \nu$ corresponding to the quantization of the flux in units of $hc/2e$.

In the above, it was assumed that the magnetic field vanishes in the entire superconductor. As is well known,² a more detailed treatment may be obtained by writing the expression for the current in the presence of an arbitrary magnetic field

$$\mathbf{i} = \mathbf{i}_s - (ne^2/\mu c)\mathbf{F}(\mathbf{A}), \quad (19)$$

where \mathbf{i}_s is the supercurrent which is not itself affected by the magnetic field and where \mathbf{F} is a nonlocal function of \mathbf{A} which reduces to $\mathbf{F}(\mathbf{A}) \approx \mathbf{A}$ for vector potentials approximately constant over distances of the order of the coherence length. By combining (19) in the usual manner with the equation

$$\nabla \times \mathbf{H} = -\Delta \mathbf{A} = 4\pi \mathbf{i}/c, \quad (20)$$

the magnetic field for given \mathbf{i}_s and external sources can be determined. Thus, by means of (2), one obtains from (19) and (20)

$$\Delta \mathbf{A} = \frac{4\pi ne^2}{\mu c^2} \mathbf{F}\left(\mathbf{A} - \frac{\hbar c}{e} \nabla \chi\right), \quad (21)$$

since χ will usually be a slowly varying function. For finite temperatures, the current associated with the back flow does not change the form of (21), but only implies a modified value for the effective number of superconducting electrons. It is seen that (21) describes a vector potential which in the interior of the super-

conductor approaches the value $(\hbar c/e)\nabla \chi$ corresponding to a flux of

$$\Phi = \oint \mathbf{A} \cdot d\mathbf{s} = -\nu. \quad (22)$$

Thus, if the dimensions of the cylinder exceed the penetration depth, the quantum states will be associated with a trapped flux (22).¹¹ The energy of the state with supercurrent ν is then, to a first approximation, that contained in the magnetic field, i.e.,

$$E_\nu = \frac{1}{8\pi} \frac{L}{\pi R_1^2} \left(\frac{\hbar c}{e}\right)^2 \nu^2, \quad (23)$$

in the limit of $L \gg R_1$.

We have based the considerations on the approximations of constant electron density. A more detailed treatment, taking into account the spatial variation of the self-consistent field, would modify some of the quantitative results, such as the expressions (12) and (10) for the angular momentum and energy of the supercurrents in the absence of magnetic effects. However, the magnetic self-coupling simplifies the situation; to the extent to which the surface effects in the penetration layer can be neglected, the wave functions (11) become exact solutions. In fact, under such conditions, the phase factor involving the velocity potential is just compensated by the magnetic coupling represented by the substitution $\mathbf{p} \rightarrow \mathbf{p} - (e/c)\mathbf{A}$ with $\mathbf{A} = (\hbar c/e)\nabla \chi$. Hence a self-consistent solution with no current in the interior of the metal is obtained if χ is chosen such that $\nabla \cdot \mathbf{A} = 0 = \Delta \chi$ and such that the wave function is univalued. This leads immediately to the states with integral values of ν ; those with half-integral values of ν are obtained, as discussed above, by at the same time shifting the single-electron states to half-integral values of the quantum number m .

The states with the persistent currents are prepared by subjecting the sample to an external longitudinal field before cooling. After the temperature has been reduced below the transition temperature, the system chooses the phase ν which minimizes the total energy in the presence of the external field, i.e., ν equal to the half-integral or integral number nearest to σ , where σ measures the number of flux units of the external field passing through the tunnel. After the external field is removed, this state is no longer the lowest state, but remains essentially stable (see Fig. 1).

The structure of the vortex lines in simply connected pieces of metal are somewhat modified by the magnetic coupling effects. As discussed above, the centrifugal energy is small compared to the magnetic field energy, and thus the critical radius r_c is determined by the

⁸ Y. Aharonov and D. Bohm, Phys. Rev. **115**, 485 (1959).

⁹ The arguments in reference 3 for important corrections to the wave function (11) in the presence of the magnetic flux would thus seem to apply only to the case of a Fermi gas without pairing correlations.

¹⁰ The energy spectrum (16) is periodic in σ , as required by the theorem of reference 3; however, as the magnetic flux is varied, the quantum number ν remains a constant of the motion and the properties of the system under such circumstances would not be periodic in σ . Thus, in a thermodynamic analysis, one should consider the grand partition function for each phase ν separately.

¹¹ Note added in proof. Recent articles [J. Bardeen, Phys. Rev. Letters **7**, 162 (1961) and H. Lipkin (preprint)] consider the corrections to the trapped flux units appropriate to systems with dimensions comparable to the penetration depth. The circulation remains quantized, but the number of electrons in such situations is not sufficient to generate the full flux (22).

balance between the field energy and the energy difference between the normal and superconducting phases. This condition implies that the field strength inside the vortex is always equal to the critical field, and $r_c^2 \sim \nu \lambda_L \xi_0$, where λ_L is the London penetration depth ($\lambda_L^{-2} = 4\pi n e^2 / \mu c^2$). We have here neglected surface effects which may be significant for small ν . Such vortex lines cannot be produced by the usual method by which the flux is trapped in a multiply connected domain, since the external field required to lower these states below the field-free state is equal to or greater than the critical field. A method for producing these states would be to first trap the flux in a multiply connected domain, in which the core is filled with another metal with a lower critical temperature. A

further lowering of the temperature will thus make the sample simply connected and the trapped flux is expected to go into one or a number of vortex lines.¹²

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¹² *Note added in proof.* Observations of trapped flux in simply connected superconductors have been made in connection with studies of intermediate state phenomena [see, e.g., D. M. Bala-shova and D. V. Sharvin, *Soviet Physics—JETP* **4**, 54 (1957); A. L. Schawlow and G. E. Devlin, *Phys. Rev.* **110**, 1011 (1958); W. de Sorbe and W. A. Healy, G. E. Report No. 61-RL-274314, July, 1961.] We are indebted to Dr. Morits, Dr. Nielsen, and Dr. Radhakrishna for calling these experiments to our attention and for interesting discussions of their significance in relation to the above considerations.

Interband Transitions for Metals in a Magnetic Field

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A quantum-mechanical derivation of the frequency and magnetic field dependence of the optical reflection and transmission in metals is given. Both interband and intraband direct transitions are considered and explicit results are obtained for a model of two simple parabolic bands having energy extrema at $\mathbf{k}=0$. Spin splitting is neglected. The calculated line shape is in good agreement with the magnetoreflection experiment of Brown *et al.* in bismuth. The results for the limiting cases of zero interband coupling or of zero magnetic field are in agreement with previous work. The study of the interband transitions in a magnetic field is shown to yield valuable information on the band structure of metals. The advantage of this method over magnetoplasma and zero-field interband studies is discussed.

I. INTRODUCTION

THE study of electronic transitions between the valence and conduction bands has yielded valuable information on the energy band structure of semiconductors.¹⁻⁵ It is of interest to ask whether similar studies can provide information on the band structure of metals.

There are two basic differences between a metal and a semiconductor, which affect the optical properties. Since, in a metal there are occupied states in the conduction band, the interband transitions are dependent on the position of the Fermi energy in the conduction band. Furthermore, the ordinary conduction processes of the "free carriers" in a good metal give rise to a background absorption, which is generally much larger than the interband effects.

This study was motivated by the observation of a resonant phenomenon in the reflection of infrared radiation from the surface of a single crystal of bismuth as the magnetic field was increased.⁶ This resonant phenomenon was associated with transitions from Landau levels in the valence band to Landau levels in the conduction band. The data on Bi were analyzed by analogy with the work on the semiconductors^{7,8} to yield values of the energy gap, and the effective mass and spectroscopic splitting factor for the conduction and valence bands. This calculation lends support to the method of analysis used in the interpretation of the bismuth data.

The object of this investigation is to determine the frequency and magnetic field dependence of the power reflection and transmission in metals. Particular emphasis is given to the effect of the magnetic field on the interband transitions and on the free-carrier

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¹ L. H. Hall, J. Bardeen, and F. J. Blatt, *Phys. Rev.* **95**, 559 (1954).

² W. C. Dash and R. Newman, *Phys. Rev.* **99**, 1151 (1955).

³ G. G. Macfarlane and V. Roberts, *Phys. Rev.* **97**, 1714 (1955); **98**, 1865 (1955).

⁴ R. J. Elliott, T. P. Mclean, and G. G. Macfarlane, *Proc. Phys. Soc. (London)* **72**, 553 (1958).

⁵ M. Okazaki, *Progr. Theoret. Phys. (Kyoto)* **25**, 163 (1961).

⁶ R. N. Brown, J. G. Mavroides, M. S. Dresselhaus, and B. Lax, *Phys. Rev. Letters* **5**, 243 (1960).

⁷ E. Burstein, G. S. Picus, H. A. Gebbie, and F. Blatt, *Phys. Rev.* **103**, 826(L) (1956).

⁸ S. Zwerdling, B. Lax, L. M. Roth, and K. J. Button, *Phys. Rev.* **114**, 80 (1959); L. M. Roth, B. Lax, and S. Zwerdling, *ibid.* **114**, 90 (1959).