

Resonant interactions of optical phonons with acceptor continuum states in silicon*

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We have performed and analyzed optical-absorption experiments which provide clear evidence of Breit-Wigner-Fano resonant interactions between the electronic continuum states of acceptors and localized excitations involving optical phonons in silicon. For boron, the 519-cm^{-1} zone-center phonon lies within the region of excitation to the continuum of the $p_{3/2}$ valence band, leading to an antiresonance at that wave number. In addition, antiresonances are observed in silicon doped with boron at 764 cm^{-1} , aluminum at 962 cm^{-1} , and gallium at 989 cm^{-1} . These numbers correspond to the one-phonon sidebands of the bound-to-bound $1S_{3/2} \rightarrow 2P_{3/2}$ impurity excitations, which lie within the region of excitation to the continuum of the $p_{1/2}$ valence band. Fano's theory is used to analyze these results.

I. INTRODUCTION

The valence bands of silicon and their related acceptor impurity states have been thoroughly studied both theoretically^{1,2} and experimentally.³⁻⁵ Because of this, the positions of the acceptor energy levels and their symmetries are known with great precision and the character of their wave functions is known to a considerable extent. The phonon spectra of silicon have also been determined by a number of techniques.⁶⁻⁸

The electron-phonon coupling for shallow acceptors in silicon is weak, as compared with crystals which have any degree of ionicity. This is one reason why the optical-absorption spectra of these defects can yield so much information; all of the spectral lines commonly reported are zero-phonon lines, and consequently are quite sharp. However, acceptor levels are separated by energies whose order of magnitude may be comparable with phonon energies, and thus interesting resonant effects^{4,5} may still occur even though the coupling is not strong. Such effects were first noted by Hrostowski and Kaiser⁹ in the bound-state optical-absorption spectrum of gallium in silicon, and were attributed by Onton, Fisher, and Ramdas⁴ to a resonant interaction of the excitation with a phonon. Subsequently these data have been further analyzed theoretically by Chandrasekhar, Ramdas, and Rodriguez,⁵ who also noted that a similar resonance occurs in aluminum-doped silicon. These resonances involve interaction between two classes of zeroth-order excitations: in one of these, the defect remains in its ground electronic state while one localized phonon is created, while in the other class a zero-phonon electronic excitation takes place to a bound excited state of the defect. When these excitations are at or near the same energies, their interaction can be detected by shifts and broadening of the bound-to-bound

zero-phonon lines.

A different resonant effect¹⁰ has been observed in optical studies of the decay of excitons bound to neutral donors in CdS and CdSe. In this case, the zone-center longitudinal-optical (LO) phonon energy is greater than the donor binding energy. Therefore, when the exciton decays, the final state of its one-phonon sideband is degenerate with no-phonon final states in which the donor is ionized. The data do not appear to be consistent with a simple Breit-Wigner-Fano (BWF) resonance^{11,12} between a discrete state and a continuum; rather, the results have been attributed to an interference between different LO phonons caused by the electron-phonon interaction.

A third class of electron-phonon resonance has been noted in the exciton absorption spectrum of several crystals, including CdTe. In this case the one-phonon sideband of the exciton is degenerate with the no-phonon continuum, and interference effects exist in the spectra.¹³

A fourth situation, leading to a BWF^{11,12} resonance, involves interference between one-phonon transitions and free-carrier absorption between valence-band states in heavily doped *p*-type silicon. Such effects have been observed by Raman scattering¹⁴ and a general theory¹⁵ for these non-local excitations has been developed. Several of the preceding examples are discussed in the review article by Levinson and Rashba.¹⁶

In this paper, we present data which indicate the existence of two types of resonant effects associated with the shallow acceptors in silicon, which have not been previously reported. The first of these occurs in boron-doped silicon and involves resonant interaction between no-phonon electronic excitations from the ground state to the continuum, and excitations in which one localized optical phonon is created while the acceptor remains in its ground electronic state. This is equivalent to the

effect observed in the bound-state spectra of gallium and aluminum,^{4,5,9} but occurs in the *continuum* of boron due to the relative shallowness of its ground state. The second effect, which has not been heretofore suggested in these systems, involves a resonance between the one-phonon sideband of the bound electronic excitations of the acceptor, and transitions to the no-phonon continuum. This is similar to the exciton resonance mentioned above, although it is quite different in detail. These effects appear as well-defined BWF resonances^{11,12} superimposed on the continuum absorption.

II. EXPERIMENTAL PROCEDURE

The absorption spectra were taken on two commercial double-beam spectrophotometers. For the energy region below 650 cm^{-1} , a Perkin-Elmer PE-21 instrument with a CsBr prism was used. Above 650 cm^{-1} , a Beckman IR-7 was used. The spectra were taken at 20.4 K with the cold sample in one of the beams. The absorption coefficients for the acceptor excitation spectra were determined by comparing the transmission of a doped crystal with that of an undoped crystal (run consecutively in the sample beam).

With the IR-7, a strip mirror replaced the usual second beam chopper (after passing through the sample) to avoid offset error due to the cold sample. No such modification was available for the PE-21 and the absorption coefficients calculated from measurements on it are therefore somewhat less accurate. The resolution of both instruments in the range of interest is estimated to be $\leq 5 \text{ cm}^{-1}$.

The boron- and aluminum-doped crystals were floating zone crystals containing $2 \times 10^{16}/\text{cm}^3$ of the corresponding acceptor. The gallium-doped crystal was pulled from the melt (Czochralski) and contained $8 \times 10^{15}/\text{cm}^3$ of gallium. The reference crystal was floating zone and undoped.

III. EXPERIMENTAL RESULTS

In Fig. 1, we show the absorption spectrum for the boron acceptor in the energy range beyond the onset of the continuum excitations to the $p_{3/2}$ valence-band edge. (The sharp bound-to-bound zero phonon excitations⁴ are at lower energies and only the last resolved peak of the series is visible in Fig. 1.) The most startling feature is the sharp dip at 519 cm^{-1} in the otherwise relatively smooth absorption tail. To our knowledge this is the first time this prominent feature has been reported. Shown dashed are two estimates for the expected absorption tail in the absence of this feature. This suggests that in addition to the dip at 519 cm^{-1} , a broader increase in absorption also exists to lower

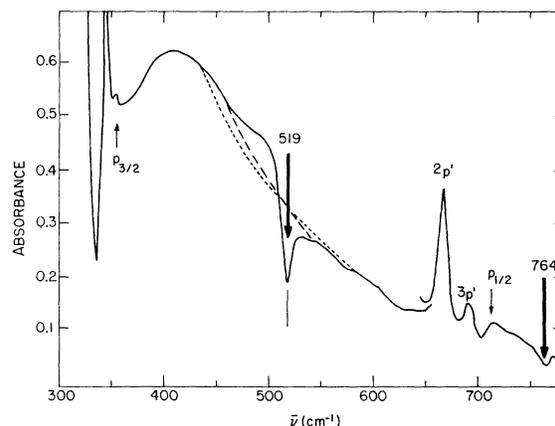


FIG. 1. Absorption spectrum at 20.4 K for the boron acceptor in silicon ($\sim 2 \times 10^{16}/\text{cm}^3$) in the energy range beyond the onset of the continuum excitations to the $p_{3/2}$ valence-band edge at 357.5 cm^{-1} .

energies.

At higher energies the zero-phonon excitations to the "bound" $2p'$ and $3p'$ states made up from the split-off $p_{1/2}$ valence band are also observed.¹⁷ Beyond the onset of the continuum excitations to the $p_{1/2}$ band, at 712.6 cm^{-1} , a second weak dip at 764 cm^{-1} is also observed.

In Fig. 2, we show that this second dip is also present and somewhat more prominent in the aluminum- and gallium-doped samples. In the figure, the spectra have been shifted so that the $1S_{3/2} \rightarrow 2P_{3/2}$ transitions at lower energies coincide. Indicated are the positions for the one-phonon (519 cm^{-1}) sidebands of these transitions.

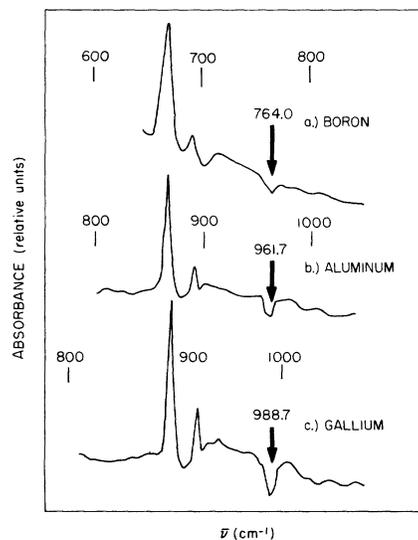


FIG. 2. Absorption spectra at 20.4 K in the region of transitions to the $p_{1/2}$ valence-band edge. The spectra have been shifted so that the $1S_{3/2} \rightarrow 2P_{3/2}$ transitions at lower energies coincide. Indicated are the positions for the one-phonon (519 cm^{-1}) sidebands of these transitions.

442.7 cm⁻¹ for aluminum, and 496.7 cm⁻¹ for gallium⁴) coincide for each. The dips are observed to occur at these wave numbers plus 519 cm⁻¹, as indicated.

IV. ANALYSIS

A. General

For both types of data presented in Sec. III the spectra appear to be characteristic of relatively weak Breit-Wigner-Fano^{11,12} resonances. The most prominent feature in both cases is a single dip (antiresonance) in a relatively strong continuum absorption. In the absence of further structure, then, it is most reasonable to begin to analyze these results by considering the mixing between a single localized state and a continuum. We shall use the form of the analysis developed by Fano.¹²

In Fano's notation, one has a Hamiltonian H , a bound state ϕ , and continuum states ψ_E . The matrix elements of H are written

$$\begin{aligned} \langle \phi | H | \phi \rangle &= E_\phi, \quad \langle \psi_{E'} | H | \phi \rangle = V_{E'}, \\ \langle \psi_{E''} | H | \psi_{E'} \rangle &= E' \delta(E'' - E'). \end{aligned} \quad (1)$$

Fano's analysis leads to an expression for the ratio of the transition probability between the initial state i and the modified continuum eigenstates Ψ_E to that between i and the unperturbed continuum ψ_E , given by

$$R(\epsilon) \equiv \frac{|\langle \Psi_E | T | i \rangle|^2}{|\langle \psi_E | T | i \rangle|^2} = \frac{(q + \epsilon)^2}{1 + \epsilon^2}, \quad (2)$$

where T is the transition operator. Here,

$$\epsilon = (E - E_\phi - F) / \frac{1}{2} \Gamma, \quad (3)$$

where

$$\Gamma = 2\pi |V_E|^2, \quad (4)$$

and

$$F(E) = P \int dE' \frac{V_{E'}}{E - E'}. \quad (5)$$

"P" means "principal part of."

The parameter q is defined by

$$\begin{aligned} q &= \left(\langle \phi | T | i \rangle + P \int dE' \frac{\langle \phi | H | \psi_{E'} \rangle \langle \psi_{E'} | T | i \rangle}{E - E'} \right) \\ &\quad \times (\pi \langle \phi | H | \psi_E \rangle \langle \psi_E | T | i \rangle)^{-1}. \end{aligned} \quad (6)$$

The magnitude of q is also given by

$$\frac{1}{2} \pi q^2 = \frac{|\langle \Phi | T | i \rangle|^2}{|\langle \psi_E | T | i \rangle|^2 \Gamma}, \quad (7)$$

where Φ is the "modified" discrete state.

The curves of R vs ϵ are asymmetric, contain-

ing the well-known resonance (increase in absorption) and antiresonance (dip in absorption). The antiresonance occurs at $\epsilon = -q$, while the resonance occurs at $\epsilon = q^{-1}$ and has magnitude

$$R_{\max} = q^2 + 1. \quad (8)$$

On the other side of the antiresonance, R is < 1 .

Consider the interaction of a single phonon with transitions to the zero-phonon defect continuum. Here ϕ is a state in which the hole is in its ground state G and one phonon is created, while for ψ_E no phonons are created and the hole is excited into the $p_{3/2}$ valence band, as modified by the boron impurity. V_E is then a measure of the electron-phonon interaction in linear coupling. We write the unperturbed states as

$$|\phi\rangle = |G1\rangle, \quad |\psi_E\rangle = |E_{3/2}0\rangle. \quad (9)$$

The transition moments in the absence of mixing are $\langle 0G | T | G1 \rangle$ for optically exciting one phonon, and $\langle 0G | T | E_{3/2}0 \rangle$ for a no-phonon transition into the continuum. V_E is given by

$$V_E = \langle 0E_{3/2} | H | G1 \rangle. \quad (10)$$

The part of H which mixes ϕ and ψ_E may be written

$$\sum_{\vec{k}_i} (Q_{\vec{k}_i}^\dagger a_{\vec{k}_i}^\dagger + Q_{\vec{k}_i} a_{\vec{k}_i}),$$

where $a_{\vec{k}_i}^\dagger$ ($a_{\vec{k}_i}$) creates (destroys) a phonon of wave vector \vec{k}_i belonging to branch i , and the operators $Q_{\vec{k}_i}^\dagger, Q_{\vec{k}_i}$ act on the hole states.⁵ Thus

$$V_E = \sum_{\vec{k}_i} \langle E_{3/2} | Q_{\vec{k}_i} | G \rangle \langle 0 | a_{\vec{k}_i} | 1 \rangle. \quad (11)$$

The form of the electronic operator $Q_{\vec{k}_i}$ depends on the details of the problem. For example, if the Fröhlich Hamiltonian¹⁸ were used, $Q_{\vec{k}_i}$ would be proportional to $e^{i\vec{k}_i \cdot \vec{r}}$.

It is clear from the experimental spectrum that a sharp antiresonance occurs at 519 cm⁻¹, preceded by a broad but discernable resonance. Analysis of the data is somewhat arbitrary, inasmuch as one must judge where the unperturbed continuum would lie; reasonable choices, however, lie between certain limits which do not strongly affect the results. Figure 3 shows the experimental curve of R versus wave number for two different choices of the unperturbed continuum, as indicated in Fig. 1.

B. One-phonon model

We first consider only one phonon (the zone-center optical phonon) in resonance with the continuum, i.e., direct application of the Fano equations.¹² The observed maximum value of R is used to determine q , Eq. (8), yielding $q \approx \pm 0.4$. The

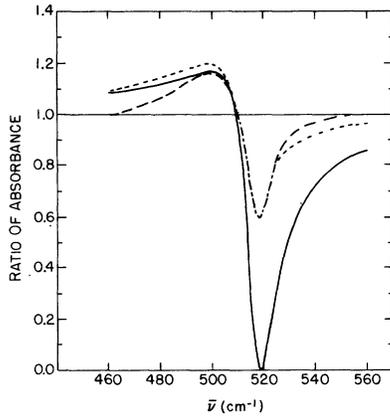


FIG. 3. Predicted Breit-Wigner-Fano resonance (solid line) for the simple case of interaction of a single lattice phonon at 519 cm^{-1} with transitions to the zero-phonon continuum of the boron acceptor in silicon. The two dashed lines represent the experimental results for two different choices for the unperturbed continuum as given in Fig. 1.

minus sign is then chosen as correct because the antiresonance is at higher energy than the resonance.

The energy scale is determined by noting that the difference between resonance and antiresonance is $q + q^{-1}$, in terms of ϵ , whereas energy scales as $\frac{1}{2}\Gamma\epsilon$. The experimental separation is 19 cm^{-1} , yielding $\Gamma = 13.4 \text{ cm}^{-1}$. E_ϕ is taken to be 519 cm^{-1} , the experimentally determined zone-center optical phonon energy.⁷ Finally, F is determined to be 2.76 cm^{-1} by the experimental result that R is a minimum at $E = 519 \text{ cm}^{-1}$, yielding, from Eq. (2),

$$R(E) = \frac{(E - 519)^2}{(E - 516.2)^2 + 44.83}, \quad (12)$$

where E is in cm^{-1} .

This theoretical curve is shown in Fig. 3. While the comparison between theory and experiment is fair, it is not outstanding. The experimental curve does not go to zero, while the theoretical curve does; and beyond 519 cm^{-1} the theoretical curve is consistently too low. While the first difference might possibly be attributable to spectrometer resolution, the second cannot be. More seriously, it is disturbing that the location of the antiresonance at the zone-center optical-phonon energy is accidental, depending on just the right value of F . A similar experimental situation occurs for the one-phonon sidebands, and it is not likely that the same accident will occur several times.

C. Distribution of phonons

We now consider a generalization of the above approach in that a distribution of phonons away

from $\vec{g}=0$ is allowed to participate in the interaction. This is a logical extension of the simpler approach, for several reasons. First, the assumption that only $\vec{g}=0$ phonons participate is valid only if the impurity wave functions are constructed from Bloch waves at $\vec{k}=0$; in reality, a range of \vec{k} space is needed for such expansions. Second, the density of available phonon states increases rapidly away from $\vec{g}=0$.

In this generalization we treat the phonons individually. We do not, for example, consider that phonons may mix through the continuum, as did Henry and Hopfield.¹⁰ This means that we simply convolve the BWF result with a normalized distribution function $h(E_\phi)$ for the participating phonons, obtaining

$$R' = \int R \left(\frac{E - E_\phi - F}{\frac{1}{2}\Gamma} \right) h(E_\phi) dE_\phi. \quad (13)$$

We have chosen to use a simple sawtooth ramp function for $h(E_\phi)$ as shown in Fig. 4, with maximum at 519 cm^{-1} . This accounts in an approximate way for the importance of the zone-center phonon while allowing other phonons to participate in a monotonically decreasing fashion.

We set $q = -1$; Fano has shown¹² that when $q^2 = 1$ and the parameters are independent of energy, there is no extra absorption due to the discrete level; it merely redistributes the continuum. For acceptors in silicon, absorption to the continuum is rather strong. On the other hand, direct transitions to the one-phonon excited states are weak (only in the presence of a defect is there a dipole moment and this is expected to be small in a non-polar material such as silicon). Therefore, $q \approx -1$ appears to be a reasonable assumption.

Equation (13) can be evaluated analytically, and by matching the depth of the resulting antiresonance and the separation between resonance and antiresonance to experiment, we determine $\Gamma = 2.8$

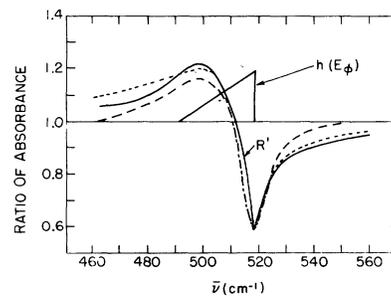


FIG. 4. Predicted Breit-Wigner-Fano resonance (solid line) for the case of interaction with a distribution of phonons as indicated. The dashed lines represent experimental results for two different choices for the unperturbed continuum as given in Fig. 1.

cm^{-1} with the width of the distribution function equal to 28 cm^{-1} , as shown in Fig. 4. The resulting predicted shape for R' now matches the data quite well, as shown. In addition, the antiresonance now occurs at 519 cm^{-1} naturally without the requirement of an accidental cancellation by F . The value of F is now zero or very small, a result of $V_E \approx \text{const}$, or small.¹²

It remains to determine whether a range of $\sim 30 \text{ cm}^{-1}$ of phonons is reasonable. This range corresponds to a distribution half-width of $\sim 15 \text{ cm}^{-1}$. From the dispersion curves of Dolling,⁶ reproduced by Temple and Hathaway,⁷ one may ascertain that this energy range corresponds to a range in phonon wave vector g of $\sim 10\%$ of $2\pi/a$, or $\sim 0.1 \text{ \AA}^{-1}$. Since the electronic wave functions G and E may be expanded in Bloch waves, the question becomes whether these states contain Bloch waves whose wave vectors k differ by $\sim 0.1 \text{ \AA}^{-1}$, since these are the functions involved in the electronic matrix elements.

The continuum states E of interest lie $\sim 63 \text{ meV}$ above the ground state, or $\sim 18 \text{ meV}$ into the valence band. Using the silicon band-structure parameters of Hensel and Feher,³ we can determine what k values correspond to this energy. These range from 0.025 to 0.06 \AA^{-1} . Since the ground state G is relatively localized, we may use the uncertainty principle to estimate the range of k values contained in it; if we assume a Bohr radius of 25 \AA , we find $k \sim 0.04 \text{ \AA}^{-1}$. This gives us a total range of uncertainty for G and E of from ~ 0.06 to $\sim 0.1 \text{ \AA}^{-1}$. This is in close enough agreement with the range of phonons used to suggest that our approach is reasonable.

We turn next to the dips observed in the continuum absorption of boron at 764 cm^{-1} , aluminum at 962 cm^{-1} , and gallium at 989 cm^{-1} . These coincide to within 1 cm^{-1} of the sum of the 519-cm^{-1} phonon and the $1S_{3/2} \rightarrow 2P_{3/2}$ bound-state excitation energy in each case, and there is thus compelling reason to attribute these features to Breit-Wigner-Fano resonances between the one-phonon sideband of the bound excitations, and the continuum. Because the experimental data contain other nearby structure and no well-defined resonances, we shall not attempt a quantitative analysis of the type done on the one phonon-continuum resonance of boron.

In this case, V_E is given by

$$V_E = \langle 0E_{1/2} | H | W1 \rangle, \quad (14)$$

where $E_{1/2}$ is the $p_{1/2}$ continuum and $|W1\rangle$ is the one-phonon sideband of excited state W . The electronic part will then be of the form $\langle E_{1/2} | Q_{\vec{g}_i} | W \rangle$.

A puzzling question is why this antiresonance is observed only in connection with the lowest excited state, especially since the intensity of the transi-

tion to this state is weaker than that to some of the other $2P$ states lying at higher energies.⁴ We have verified that there are no rigid selection rules to forbid coupling of these higher states to the continuum. We suspect that the answer lies in the mismatch of k space occupied by W and $E_{1/2}$. The excited states W are diffuse and sample much less k space than the ground state G , and the higher the states the less k space they use. The k space occupied by $E_{1/2}$ is determined by its energy, and is farther from $k=0$ as the energy increases. Whereas for the one-phonon resonance matrix element $\langle E_{3/2} | Q_{\vec{g}_i} | G \rangle$, one had good overlap of k space among all factors, in the present case the overlap is small for the phonons of interest and becomes even smaller for resonances involving higher excited states, in which $E_{1/2}$ samples larger values of k and W samples smaller values. The antiresonance should be there, but more difficult to observe.

Figure 5 shows a composite of results for the one-phonon antiresonance and the one-phonon-sideband antiresonance for boron in silicon. The widths of the bound states indicate their approximate range in k space.

V. DISCUSSION AND CONCLUSIONS

The preceding sections have summarized the experimental results and have provided theoretical arguments indicating that Breit-Wigner-Fano resonant interactions have been observed. In the case

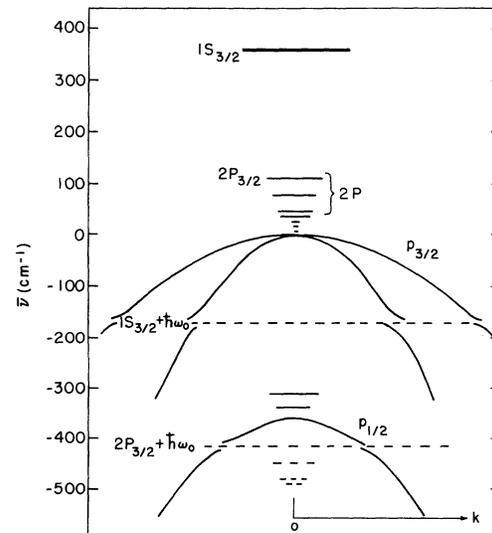


FIG. 5. Schematic illustration of the acceptor states for boron in silicon, the valence-band states, and the resonant interactions. The width of the bound states indicates their approximate range in k space.

analyzed in greatest detail, one localized phonon degenerate with excitations from the ground state of the boron impurity to its continuum states, the Fano parameter "q" was found to be negative. We may analyze Eq. (6) to ascertain whether a negative sign for q is understandable.

The argument given earlier that $\langle \phi | T | i \rangle$ is small (or zero) means that we may write Eq. (6) as

$$q \cong \frac{1}{\pi} P \int dE' \left(\frac{\langle \phi | H | \psi_{E'} \rangle \langle \psi_{E'} | T | i \rangle}{\langle \phi | H | \psi_E \rangle \langle \psi_E | T | i \rangle} \right) \frac{1}{E - E'}. \quad (15)$$

q [actually, $q(E)$] will be positive if the contribution to the integral for $E' < E$ exceeds that for $E < E'$, while it will be negative if the contribution for $E < E'$ exceeds that for $E' < E$.

Experimentally, the continuum falls off with increasing energy in the region of interest; therefore, the contribution from $\langle \psi_{E'} | T | i \rangle / \langle \psi_E | T | i \rangle$ will be larger for $E' < E$ than for $E' > E$. This is analogous to a situation analyzed by Fano¹² and alone would make q positive. Whether the ratio $\langle \phi | H | \psi_{E'} \rangle / \langle \phi | H | \psi_E \rangle$ can increase with increasing E' , and faster than the other ratio falls off, is difficult to predict; in one model we have found F [Eq. (5)] to be zero, which means that this ratio is constant in E' ; in another we have found F to be positive, meaning that this ratio decreases with in-

creasing E' . Fano, in analyzing Eq. (15) for a different situation, treated this ratio as constant in E' . There is then no simple indication that the q of Eq. (15) should be negative.

An alternative explanation is that $\langle \phi | T | i \rangle$ in Eq. (6) might be both negative and large enough to make q negative. If so, then the value $q = -1$ which we have chosen is accidental.

More information could be brought to bear on this question and others if higher-resolution data were obtained. Not only could the boron phonon resonance be studied with greater precision, but further analysis of the one-phonon sideband resonances would be possible and other resonances from other bound states might be detected.

We may nevertheless conclude that interactions between zone-center optical phonons and the continuum states of shallow acceptors in silicon have been observed for the first time in excitation spectra. This type of study may provide a powerful and direct method of studying the vibronic coupling in such systems.

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¹Many papers on the band structure of silicon have appeared. See, e.g., M. Cardona and F. H. Pollak, *Phys. Rev.* **142**, 530 (1966).

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