jection has been found as a result of the electromagnetic cyclotron instability driven by $T_{i\perp} > T_{i\parallel}$.[16]

The analysis may also explain why apparently no anomalous beam diffusion has been observed to date in parallel-injection experiments. Figure 1 shows that ion-beam instabilities require injection velocities $V_D \sim C_A$, but recent experiments conducted at low $\beta \lesssim 0.005$ have used $V_D \sim 10 V_p \lesssim C_A/4$. Instability thus requires a toroidal ion β of about 0.05, far in excess of the presently attainable values in tokamaks.

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Two-Phonon Spectrum of Diamond*†

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A careful calculation of the one-phonon dispersion in diamond reveals a shallow maximum in the [100] direction for the Δ_2' (LO) branch. The two-phonon (overtone) density of states shows a sharp peak near twice the Raman frequency. This supports the interpretation of an observed peak in two-phonon Raman scattering as a simple overtone without the need to invoke a two-phonon bound state.

The recent careful experimental investigation of the Raman spectrum of diamond[1] has stimulated considerable theoretical work attempting to explain the observed spectral features. Particular attention has been given to the origin of the sharp, polarized peak in the Stokes Raman scattering at an energy shift slightly above twice the one-phonon zone-center phonon energy and with symmetry $\Gamma^{(1+)}$. It has been proposed that this peak represents a two-phonon bound state.[2] In the present work we present new theoretical results which give strong support to interpreting this feature as an ordinary two-phonon overtone, arising from novel phonon dispersion in diamond.

The essential point of our work has been a very careful investigation of the phonon dispersion in diamond using a fine grid for calculating $\omega(\vec{q})$. We used a valence-force-field model[3] which gives excellent agreement with the $\omega(\vec{q})$ determined from inelastic neutron scattering. The same model was also successful (with different parameters) in investigating phonon dispersion in silicon and germanium.[3] In Fig. 1 we show phonon dispersion in diamond in the direction $\vec{q} \parallel \Delta[100]$. The calculated points are shown. To be noted is the shallow maximum in the branch $\Delta_2'(0)$ at q approximately $\frac{1}{3}$ of the zone boundary (X point). This maximum in this branch is approximately

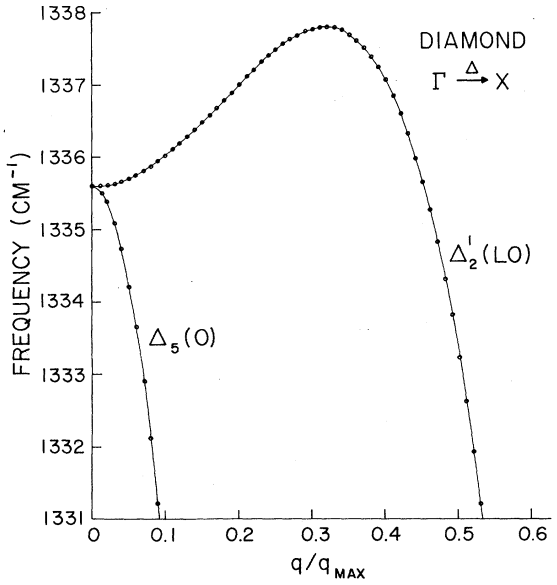

FIG. 1. Details of dispersion curves for diamond around Γ in [100] symmetry direction.

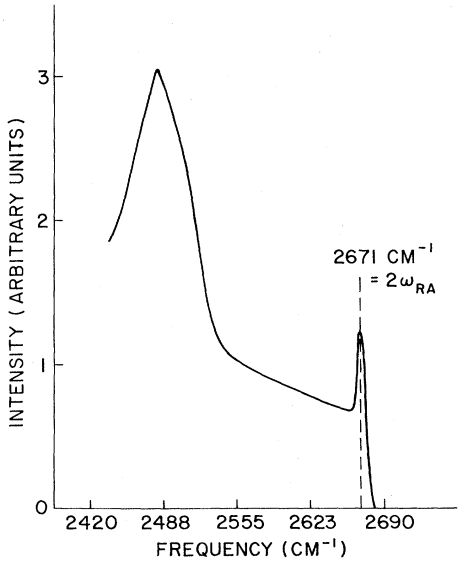

FIG. 2. Overtone density of states (high-frequency section) for diamond. Resolution of the calculation 2.7 cm^{-1}.

2.2 cm^{-1} *above* the local Γ calculated value: 1335.6 cm^{-1}. It was entirely missed in previous calculations, and no neutron scattering data exist near this energy-momentum transfer on this branch. To be noted is that for all other branches [$\Delta_5(0)$ shown, but also in other directions: Λ and Σ] Γ is a maximum. Under Phillips's[4] scheme the point Γ in diamond is a fluted critical point of type "F_2," while the point $\vec{q} \approx 0.3\vec{X}$, where the LO branch has its maximum on the Δ line, is a P_3 critical point (absolute maximum).

The two-phonon overtone density of states

$$\rho_2(\omega) = \sum_j' \sum_{\vec{q}} \{\delta(\omega) - [\omega_j(\vec{q}) + \omega_j(-\vec{q})]\},$$

where j is a branch index, can be directly computed from our results. In Fig. 2 this is shown for diamond, in the region of twice the one-phonon Raman frequency. The sharp feature at 2671 cm^{-1} (i.e., twice the one-phonon Raman frequency) arises from the F_2 critical point at Γ superimposed on the background. The width of this feature is about 5–6 cm^{-1}, and its relative intensity is about $\frac{1}{3}$ that of the two-phonon feature at about 2450 cm^{-1}. The relative intensity is in reasonable accord with experimental results; the width of the feature is in accord with a recent preliminary remeasurement by Washington.[5] The center frequency of the peak is not displaced from $2\omega_{RA}$ in our calculation since it arises from the F_2 singularity in $\rho_2(\omega)$ at Γ: Presumably inclusion of anharmonicity will produce a slight shift (2 cm^{-1} in 2671 cm^{-1}) in the real part of the overtone self-energy and is now being investigated.

To be convinced that this result is not an artifact, the same calculation was performed for silicon. As expected in silicon Γ is a P_3 point in all branches. The two-phonon overtone density of states was computed for silicon and is shown in Fig. 3. Near the two-phonon cutoff $\rho_2(\omega)$ decreases smoothly to zero as $(2\omega_{RA} - \omega)^{1/2}$. This is in accord with the absence of any experimental indication of a peak at $2\omega_{RA}$ in Raman scattering in silicon.[6]

A tentative understanding of the origin of the LO branch dispersion along Δ can be obtained from a Born force-constant model[7] using interactions up to the second neighbor. The secular determinant factorizes for this branch in this direction and the condition for a maximum is $8\mu/\alpha > 1$ (α and μ being first- and second-neighbor force constants, respectively). Transforming the valence-force constants of our model into Born's parameters we find $8\mu/\alpha = 1.57$ (diamond), 0.66 (silicon), 0.87 (germanium). Since the second-neighbor force constant μ can be related to angle bending and its interactions with other internal coordinates it appears that the higher ratio of μ/α reflects a greater angular character of forces in diamond than in silicon and germanium. General chemical intuition would support this view.

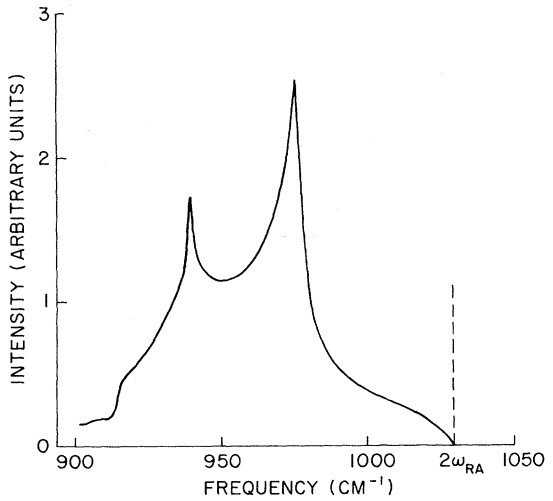

FIG. 3. Overtone density of states (high-frequency section) for silicon. Resolution of the calculation 1 cm^{-1}.

The reversal of the order of phonon branches at X (X_1 above X_4 in diamond, the reverse in silicon and germanium) also is a consequence of the greater relative bond-angle stiffness.

We believe the proposed interpretation of the peak near $2\omega_{RA}$ in diamond as an overtone has several merits in addition to its simplicity. It resolves the puzzle as to why diamond, which is less anharmonic than silicon or germanium, would have a bound state while the latter two did not. Also the magnitude of the anharmonic coupling needed in diamond for a two-phonon bound state to occur is some 5–10 times larger than would be consistent with other properties which also reflect anharmonicity (such as thermal expansion).[8] The temperature dependence of the frequency and shape of the peak is omitted from our harmonic theory. However, inclusion of anharmonic terms are expected to produce these effects.

Apparently the first suggestion that the LO branch in diamond might have its absolute maximum at $q \neq \Gamma$ was advanced by Uchinokura, Sekini, and Matsuura.[9] Later calculations of Raman scattering by Go, Bilz, and Cardona[10] and by Tubino and Piseri[11] using a bond-polarizability model could be interpreted as indicating either that the relevant matrix element was very rapidly increasing near $2\omega_{RA}$, or else that the diamond phonon dispersion was unconventional. Our calculation gives strong support to the latter and it would be desirable to verify this prediction directly, e.g., by neutron scattering.[12]

Comparison of the results of our calculation, Fig. 2, with the published experimental results (Fig. 1 of Ref. 1, and Ref. 5) support the interpretation of the peak near $2\omega_{RA}$ as originating from a simple overtone without the need to invoke a two-phonon bound state. In order fully to confirm this interpretation, calculation of the peak shift, line shape, and temperature dependence would be desirable. This work is in progress.[13]

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