Quasielastic light scattering in silicon

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A dominant, broad central mode in the Brillouin spectrum of silicon was first reported by Sandercock in 1978. This feature was later ascribed to a two-phonon difference scattering process. However, the issue of quasielastic scattering in silicon has remained a major outstanding problem in Brillouin scattering, because similar central modes in other materials have commonly displayed two components. A second, broader quasielastic mode has been measured and is reported here.

I. INTRODUCTION

Quasielastic light scattering, leading to central modes in the Brillouin spectrum of dielectrics, has attracted considerable interest due to its relationship with the critical behavior of the Rayleigh peak near second-order phase transitions.1,2 The scattering may arise from a variety of mechanisms comprehensively reviewed by Bruce and Cowley.3 The presence of quasielastic scattering in silicon has been the subject of controversy since it was first observed by Sandercock in 1978.4 In Fig. 1 the scattering can be seen as a broad central mode in the Brillouin spectrum. Although a significant quasielastic peak due to scattering from entropy fluctuations had been predicted by Wehner and Klein,5 the central mode shown in Fig. 1 does not match the width expected from the decay rate of these diffuse excitations. Furthermore, as discussed below, a nearly collision free regime prevails in silicon at room temperature, making an entropy fluctuation model inappropriate. Detailed studies by Anderson et al.6 subsequently suggested that the observed central mode [full width at half maximum (FWHM) ~60 GHz] arises through difference scattering from pairs of optic phonons on the same dispersion branch.

Central mode scattering with two components has been reported in several transparent materials, including KTaO3,7 SrTiO3,8 diamond,9 and TiO2.10 The work in Refs. 9 and 10 suggests that in the collision free regime the two components probably arise from two-phonon difference scattering from a single acoustic phonon branch and from different phonon branches, respectively. In that case, it remained mysterious that only a single mode was observed in silicon.

In the present paper we report the observation of a second quasielastic mode in silicon. The probable origin of this mode is discussed in relation to existing theories.

II. THEORETICAL DISCUSSION

Brillouin scattering arises from strain-dependent fluctuations in the permittivity ε caused by bulk acoustic phonons. In fact, ε may also be influenced by variations in temperature (or equivalently entropy), resulting from the collective motion of the gas of thermally excited phonons. In terms of phonon-transport theory, such collective motion can be described by nonpropagating variations in the local phonon number density. These fluctuations are a meaningful concept only in the hydrodynamic regime where the scattering phonon q makes many collisions in a distance 1/q, and so temperature and entropy can be defined on these scales. Therefore entropy fluctuations normally obey the law of heat diffusion and lead to scattering at the same central frequency as the incident radiation. The angular frequency width of the peak Γ (FWHM) is determined by the decay rate of the scattering fluctuations. Thus Γ=2Dthq2, where Dth is the thermal diffusivity.

The ratio of the scattered intensity in the central peak IC to that in the Brillouin peak IB is given by the Landau-Placzek ratio IC/2IB=(Cp/Cv)−1=RLP, where Cp (Cv) is the specific heat per lattice site at constant pressure (volume). Although RLP is small in solids because Cp≈Cv, Wehner and Klein5 showed that the ratio must be modified by a solid-state enhancement factor to allow for the full effect of entropy fluctuations on the dielectric constant. The enhancement factor was evaluated in Ref. 5 for a range of materials and it was predicted that silicon would show the greatest enhancement for scattering from entropy fluctuations among the materials considered.

However, phonon-transport theory is no longer valid in the regime of high frequency and low temperature, since the phonons make few collisions and entropy cannot be defined locally. In the case of silicon at room temperature, the pho-

FIG. 1. The Brillouin spectrum of silicon displays a broad quasielastic peak around the elastically scattered central peak. The latter has been removed here due to distortions introduced by a shutter system used to protect the photomultiplier tube from saturating. Peaks corresponding to the surface acoustic wave (S), bulk transverse mode (T), and longitudinal mode (L) can also be observed.
non mean free path $\bar{T}$ can be estimated by $3D_{th}/\bar{v}$, where $\bar{v}$ is an average phonon velocity. Using data from Ref. 11, this gives $lq/2\pi \approx 0.8$, which indicates a nearly collision free regime. Nevertheless, scattering may still occur via the second-order Raman effect arising from two-phonon difference processes, where a phonon $Q$ (frequency $\Omega$) is created with the simultaneous annihilation of a phonon $Q-q (\Omega - \omega)$. Recent work on TiO$_2$ suggests that two-phonon difference processes could occur between two different phonon branches or from a single branch, leading to a two-component peak. In contrast to the $q^2$ dependence of scattering from entropy fluctuations, it is noted that two-phonon difference processes involving the same branch should scale proportionally to $q$, while processes involving different phonon branches are expected to be independent of $q$.

The above concepts can be illustrated by diamond, where a central mode with two components has been observed. Two-phonon difference processes were invoked to account for the two components at low temperatures. The narrow component exhibits a $q$-dependent linewidth, while the broad component does not depend on $q$. At higher temperatures, where the phonons undergo many collisions, the linewidth of the narrow component was observed to become equal to that predicted for scattering from entropy fluctuations.

In contrast to the various transparent materials discussed above, only a single central component, with FWHM $\sim 60$ GHz, has previously been observed in the silicon spectrum. The dependence of the quasielastic spectrum of silicon on temperature, doping level, wave vector, polarization, and intensity of the incident light has been studied in detail by Anderson et al. Their results were substantially in accord with the behavior predicted for two-phonon difference scattering from a single branch, leaving the absence of the second component a topic of some controversy.

**III. EXPERIMENTAL RESULTS**

It should be noted that the broader central mode component typically extends to frequency shifts beyond the Brillouin peaks. Moreover, silicon is relatively opaque for visible wavelengths and so the scattering volume is restricted to a relatively narrow surface layer. A low intensity peak of this width would be difficult to observe on the scale of the spectrum shown in Fig. 1, so measurements were performed with a Sandercock-type tandem Fabry-Perot interferometer set to a relatively large free spectral range of 600 GHz.

Reflection losses of the incident beam were reduced by polarizing the beam in the sagittal plane and a large aperture lens ($f/2.3$) was used to improve collection efficiency. Nevertheless, these low intensity spectra required accumulation times of up to 12 h. The scattered light passing through the interferometer was detected by a cooled photomultiplier tube with less than one dark count per second. The silicon sample had a relatively low doping level of $10^{16}$ cm$^{-3}$, which suggests that free-carrier excitations would not play any role in the light scattering.

Three wavelengths of an argon-ion laser operated in a single mode were used in order to study the $q$ dependence of the scattered light. For a backscattering geometry, the transferred wave vector is given by $q = 2nk$, where $k$ is the wave vector of the incident beam and $n$ is the refractive index. Therefore the transferred wave vector increases by about 14.4% on going from the 514.5 nm laser line to 476.5 nm.

The resulting spectra display a broad low intensity peak, as shown in Fig. 2, scaled to the height of the longitudinal bulk mode ($L$). The Rayleigh ($R$) and bulk transverse ($T$) modes can also be seen, but the surface wave and Lamb shoulder are not resolved here. The increase in the longitudinal phonon frequency is consistent with the increase in the wave vector described above. In attempting to estimate the width of this central peak, one has to allow for the longitudinal and transverse bulk peaks as well as the intense elastic peak at the laser frequency. The latter clearly includes contributions to its tails from the narrow quasielastic mode as well as the ripple scattering. Unfortunately, it is impractical to subtract all these various contributions from the spectrum. Instead it was decided to exclude the longitudinal bulk peaks and all the channels between and including the transverse bulk peaks. This was motivated by Fig. 1, which suggests that the narrow central mode does not extend significantly past the transverse peak.

A Gaussian function with a constant background was fitted to the remnants of each spectrum, as shown in Fig. 3. The fitted curves suggest that the central peak has a width of 300–400 GHz. Note that the peaks are slightly asymmetric, leading to a displacement of the fitted curves toward negative frequency shifts. Although both Lorentzian and Gaussian curves...
curves were sensitive to small changes in the background level, it was found that the Gaussian curves provided a more reasonable estimation of the width and background.

IV. CONCLUSION

A second quasielastic light scattering component has been observed in silicon. Given the difficulty of fitting the observed mode, it is not possible to determine the \( q \) dependence of this mode with certainty. However, the results appear to be more consistent with the \( q \) independence of two-phonon difference scattering involving different phonon branches. In any event, difference scattering from a single branch cannot scatter light with a frequency shift larger than the Brillouin component.\(^{10}\) Although the linewidth of the additional mode is comparable to that predicted for scattering from entropy fluctuations, this appears to be coincidental, as the present sample falls into an almost collision free regime at room temperature. The low intensity of the peak explains why it was not previously observed.

It should be possible to confirm the origins of this mode by studying the temperature dependence of the width and by comparing theoretically predicted line shapes based on the phonon dispersion of silicon.

The observation of this broad central mode removes an anomaly: silicon displays a two-component low frequency spectrum that is similar to that observed in transparent solids with measurable quasielastic scattering.

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