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# Lattice isotope effects on the widths of optical transitions in silicon

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## Abstract

Using measured isotope shifts for the  $607\text{ cm}^{-1}$  local vibrational mode, LVM, of substitutional carbon,  $C_s$ , we demonstrate that isotope disorder contributes  $\sim 0.5\text{ cm}^{-1}$  to the width of that LVM in natural silicon. The width of the LVM of  $C_s$  also depends on its energy relative to the density of two-phonon states, and increases along the sequence  $^{13}\text{C}$  in natural silicon,  $^{12}\text{C}$  in natural silicon and  $^{12}\text{C}$  in  $^{30}\text{Si}$ . Other LVMS show larger isotope effects, and so discrete structure rather than line broadening. In the case of zero-phonon lines, we take the  $3942\text{ cm}^{-1}$  line as a potentially favourable example. We estimate that the isotope disorder contributes only  $0.09\text{ cm}^{-1}$  to the linewidth in a natural silicon sample, a contribution that is negligible compared to typical strain-broadening effects, but would be a lower limit to the linewidth in high-purity natural silicon.

## 1. Introduction

Recent work on single-isotope silicon has allowed macroscopic properties to be investigated, such as self-diffusion (Bracht *et al* 2005) and the effect on the lowest energy gap of changing the isotopes (Karaiskaj *et al* 2002, Hayama *et al* 2004b). There has also been considerable interest in the microscopic properties of optical centres. For example, the effects have been reported of isotope substitution on the local vibrational modes of hydrogen-related centres (Pereira *et al* 2003) and of interstitial oxygen (Kato *et al* 2003). We have recently reported the isotope shifts of 'deep' optical centres, with transition energies between 760 and 970 meV, which have been generated by electron-radiation damage or by ion implantation (Hayama *et al* 2004b, 2004a). One surprising result has been the discovery that a well-defined ground-state splitting of acceptor energy levels is caused by the random isotope content of natural silicon (Karaiskaj *et al* 2003a).

Isotopic disorder in natural silicon has also been shown to broaden the photoluminescence arising from excitons bound to shallow donors and acceptors (Karaiskaj *et al* 2001). The absolute value of the broadening is small, at  $\sim 0.04 \text{ cm}^{-1}$ , since the excited state samples of the order of  $10^4$  silicon atoms, and so the disorder tends to average out. Nevertheless, the broadening is sufficient enough that some optical transitions at acceptor centres in natural silicon are not resolved (Karaiskaj *et al* 2001).

Isotope disorder effects will be more significant at localized centres. In this paper we enquire to what extent the disorder will influence optical transitions at these localized centres. We will show that isotope disorder can be detected in local vibrational modes, but its effects are negligible compared to strains for zero-phonon lines of deep centres.

## 2. Technical aspects

The absorption measurements were made with the samples at 8 K on the cold finger of a closed cycle cryostat, using a Nicolet Fourier transform spectrometer fitted with a KBr beam splitter and an MCT detector cooled to 77 K. The resolution, as given by the full width at half height of residual atmospheric absorption, was  $0.5 \text{ cm}^{-1}$ . For clarity in the figures, only one in three measured datum points are shown in the experimental spectra, and spectra have been displaced vertically where necessary.

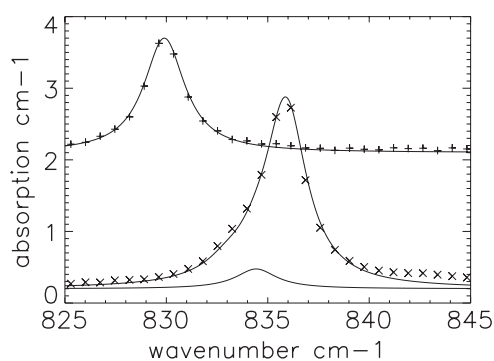
Isotope effects have been measured using a sample of isotopically enriched Czochralski-grown  $^{30}\text{Si}$  with abundances  $^{28}\text{Si}$  (0.67%),  $^{29}\text{Si}$  (0.59%) and  $^{30}\text{Si}$  (98.74%), giving a mean mass number of  $\overline{M}(^{30}\text{Si}) = 29.98$ . The sample contained substitutional carbon at a concentration of  $[\text{C}] \sim 3 \times 10^{17} \text{ cm}^{-3}$ , interstitial oxygen at  $[\text{O}] = 8.8 \times 10^{17} \text{ cm}^{-3}$  and substitutional boron at  $[\text{B}] = 5.5 \times 10^{16} \text{ cm}^{-3}$ . For data from natural-abundance silicon, a Czochralski sample with similar  $^{12}\text{C}$  and  $^{16}\text{O}$  concentrations was used, plus a Czochralski sample doped with  $^{13}\text{C}$ . The samples were irradiated with 1.5 MeV electrons to a dose of  $5 \times 10^{17} \text{ cm}^{-2}$  at nominal room temperature on a water-cooled copper block. A high-purity float-zone sample has been used as a reference sample for the absorption spectra.

Density functional theory has been used to calculate the frequencies of local vibrational modes of interstitial oxygen, the vacancy–oxygen centre and substitutional carbon. Dynamical matrix elements were evaluated using linear response theory, as in Pruneda *et al* (2002), or by explicitly displacing the impurity and its silicon neighbours along all the Cartesian axes as in Coutinho *et al* (2001).

## 3. Local mode effects

A local vibrational mode (LVM) is severely affected by the mass of the immediate neighbours of the light impurity atom. As an example, the interstitial oxygen atom  $\text{O}_i$  bonds to two Si neighbours. We calculate that if all the Si atoms are changed from  $^{28}\text{Si}$  to  $^{30}\text{Si}$ , the  $1136 \text{ cm}^{-1}$  mode will change in energy by  $-7.1 \text{ cm}^{-1}$  (Hao *et al* 2004), in close agreement with the  $-7.3 \text{ cm}^{-1}$  of experiment (Kato *et al* 2003). This shift is produced primarily by the two immediate neighbours. We calculate that if only one of the six next-neighbours is changed, then the shift is  $-0.04 \text{ cm}^{-1}$ , a negligible contribution to the total shift. The large isotope shift, and its dependence on the two immediate neighbours of the  $\text{O}_i$  atom, result in the well-known discrete isotope structure of the  $1136 \text{ cm}^{-1}$  LVM (Kato *et al* 2003).

The vacancy–oxygen ‘A’ centre is another example of an oxygen atom primarily bonded to two Si atoms, in a structure we represent by Si–O–Si. In the neutral charge state its isotope dependence is as shown in figure 1. In  $^{30}\text{Si}$ , the line is symmetric and Lorentzian in shape

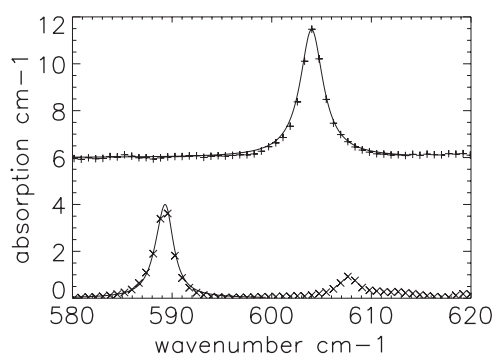


**Figure 1.** The absorption of the vacancy–oxygen centre in  $^{30}\text{Si}$  (plus signs, +, displaced vertically for clarity) and in natural silicon (crosses,  $\times$ ). The line through the  $^{30}\text{Si}$  data is a single Lorentzian of full width at half height  $\Gamma = 2.45 \text{ cm}^{-1}$ . The line through the data for the natural silicon is a sum of Lorentzians of the same width,  $2.45 \text{ cm}^{-1}$ , weighted according to the random isotope content of two equivalent silicon sites. The contribution from one  $^{29}\text{Si}$  and one  $^{28}\text{Si}$  is shown explicitly.

with a full width at half height of  $\Gamma = 2.45 \text{ cm}^{-1}$ . The line is shifted by  $-5.9 \text{ cm}^{-1}$  relative to the line in natural silicon. We have repeated the methods detailed in Markevitch *et al* (2004) with changes in the mass-weighted dynamical matrix. We predict that in  $^{30}\text{Si}$  the line would be shifted by  $-6.4 \text{ cm}^{-1}$ , in good agreement with experiment. If only one immediate neighbour is  $^{30}\text{Si}$ , giving  $^{30}\text{Si}-^{16}\text{O}-^{28}\text{Si}$ , the shift is predicted to be almost half, at  $-3.1 \text{ cm}^{-1}$ , while replacing both the bonded Si atoms, giving  $^{30}\text{Si}-^{16}\text{O}-^{30}\text{Si}$ , produces a shift of  $-6.4 \text{ cm}^{-1}$ . Since the isotope effect is determined by the two nearest-neighbour Si atoms, the lineshape may be calculated by a sum of six Lorentzians, each weighted according to the relative probability of having each combination of randomly chosen isotopes at the two sites. We use the *same width* of  $\Gamma = 2.45 \text{ cm}^{-1}$  as for  $^{30}\text{Si}$ , and the position of each peak is determined by the simple rule that one  $^{30}\text{Si}$  and one  $^{28}\text{Si}$  give half the shift of two  $^{30}\text{Si}$  atoms, and each  $^{29}\text{Si}$  gives half the shift of each  $^{30}\text{Si}$  atom. The lineshape and its asymmetry are reproduced in figure 1. We show explicitly the contribution from one  $^{28}\text{Si}$  and one  $^{29}\text{Si}$ , to demonstrate that the isotope shifts are sufficiently large that they produce discrete structure, rather than a broadening effect. The lineshape in natural silicon is typical of many LVMs in that the large isotope shift results in the tendency to form discrete structure rather than broadening.

To obtain broadening without discrete structure we need a relatively small isotope shift. We have found that the substitutional carbon atom,  $\text{C}_s$ , has the smallest isotope shift of the LVMs studied so far (Davies *et al* 2005). Figure 2 shows the absorption produced by  $\text{C}_s$  in a sample of natural silicon doped with  $^{13}\text{C}$ , and a sample of  $^{30}\text{Si}$  doped with  $^{12}\text{C}$ . (The position of  $^{12}\text{C}$  in natural Si is indicated by the trace of  $^{12}\text{C}$  in the nominally  $^{13}\text{C}$  sample.) The intrinsic absorption of the silicon lattice has been removed by subtracting the spectrum of the pure reference sample. In the case of  $^{30}\text{Si}$ , the reference spectrum was scaled down in frequency by the square root of the mean atomic masses of the  $^{30}\text{Si}$  sample relative to the float-zone sample.

We note that the shift of the transition from natural silicon to  $^{30}\text{Si}$  is by  $-3.5 \text{ cm}^{-1}$ . We have calculated the dynamical matrix of a  $\text{Si}_{63}\text{C}_s$  periodic supercell from first principles, using linear response theory (Sanati *et al* 2004). The calculated isotope shift is  $-4.6 \text{ cm}^{-1}$ , in close agreement with experiment. Again, let us calculate the effect of isotope disorder. We recall that  $\text{C}_s$  is a substitutional atom, with  $T_d$  symmetry, so there are four nearest neighbours. The local vibrational mode (LVM) transition is from the zero-point state, transforming as  $A_1$ , to a triply degenerate mode  $T_2$ . If three of the neighbours of  $\text{C}_s$  were  $^{28}\text{Si}$  and one was  $^{30}\text{Si}$ , the



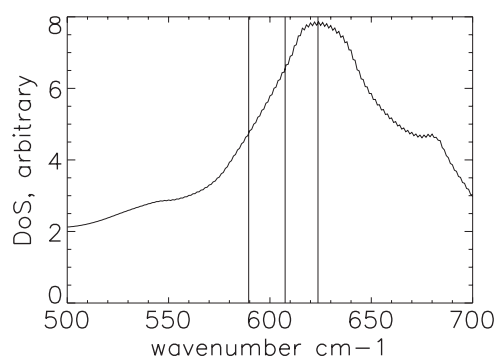
**Figure 2.** The upper spectrum, which has been displaced vertically for clarity, shows the absorption in a  $^{12}\text{C}$ ,  $^{30}\text{Si}$  sample. Plus signs (+) are measured data after subtraction of the intrinsic absorption (predicted from a pure, natural silicon sample). The line through the points + is a Lorentzian curve of width  $2.8\text{ cm}^{-1}$ . The lower spectrum shows the absorption in a  $^{13}\text{C}$ , natural silicon sample. Crosses (x) are measured data after subtraction of the intrinsic absorption (of a pure, natural silicon sample). The two peaks are produced by  $^{13}\text{C}$  and  $^{12}\text{C}$  atoms in substitutional positions. The line through the  $^{13}\text{C}$  peak is a sum of Lorentzians weighted according to the random isotope population of the four nearest-neighbour sites to the carbon atoms, each of width  $1.9\text{ cm}^{-1}$ .

lower symmetry would split the  $T_2$  level. Calculation gives predicted shifts of  $-1.0\text{ cm}^{-1}$  for the doublet and  $-1.9\text{ cm}^{-1}$  for the singlet, with a weighted mean of  $-1.3\text{ cm}^{-1}$ , essentially one quarter of the shift predicted when all the neighbours are  $^{30}\text{Si}$ . We assume that two  $^{30}\text{Si}$  atoms produce half the total shift, and three give three-quarters, and that the intrinsic lineshape of all the components is Lorentzian with equal full widths at half height of  $\Gamma$ . The simulated lineshape is shown in figure 2 for the natural silicon sample with  $\Gamma = 1.9\text{ cm}^{-1}$ . The random isotopes contribute about  $0.5\text{ cm}^{-1}$  to the width.

Although isotope disorder broadens the line, in single-isotope  $^{30}\text{Si}$  the observed linewidth is actually greater, at  $2.8\text{ cm}^{-1}$ . This is not an experimental artefact of temperature or of strains—the zero-phonon lines in this sample were sharper than for comparable natural-isotope Czochralski silicon, and we have seen that the LVM of the A-centre can be fitted in the same samples using identical linewidths in natural silicon and  $^{30}\text{Si}$ . The linewidth of most LVMs is mainly caused by the decay time. The  $C_s$  peak is superimposed on the intrinsic two-phonon absorption, suggesting that the decay is likely to be into two lattice modes. The optical absorption process is restricted to creating two phonons of equal and opposite wavevector, while wavevector has little meaning for the decay process at the highly localized  $C_s$  vibration. Figure 3 shows the density of two-phonon states for  $^{28}\text{Si}$ , calculated from the one-phonon density of Giannozzi *et al* (1991). The position of the  $C_s$  mode is marked for  $^{28}\text{Si}$  and also its effective position (the observed energy scaled by  $\sqrt{29.96/28.11}$ ) in  $^{30}\text{Si}$ . We note the 60% increase in the number of decay channels available to the LVM of  $^{12}\text{C}$  in  $^{30}\text{Si}$  compared to  $^{28}\text{Si}$ , which is at least a partial explanation of the 50% increase linewidth. The width of the LVM for  $^{12}\text{C}$  in natural silicon is  $\Gamma = 2.1\text{ cm}^{-1}$ , after removing the disorder effects, slightly greater than for  $^{13}\text{C}$ , again in qualitative agreement with the density of states picture (figure 3).

The change in linewidth of  $C_s$  by changing the lattice isotope is the corollary of the dependence of the linewidth of the  $1136\text{ cm}^{-1}$   $O_i$  line as the impurity isotope is changed (Sun *et al* 2004).

To summarize, we have demonstrated that isotope disorder effects are present for the  $C_s$  mode in natural silicon. In a 50:50 mixture of  $^{28}\text{Si}$  and  $^{30}\text{Si}$ , the isotope disorder would



**Figure 3.** Part of the two-phonon joint density of states, DoS, calculated from the one-phonon density of Giannozzi *et al* (1991). The vertical lines show, from left to right, the frequency of the  $C_s$  LVM of  $^{13}\text{C}$  in natural silicon, that of  $^{12}\text{C}$  in natural silicon, and that of  $^{12}\text{C}$  in  $^{30}\text{Si}$ . For the last, the frequency has been increased from the measured value of  $604.0\text{ cm}^{-1}$  by multiplying by  $\sqrt{29.96/28.1}$  to place the LVM at the correct location on the shape of the DoS.

contribute  $1.5\text{ cm}^{-1}$  to the width of the LVM of  $C_s$ . However, as a result of the change in position of the LVM relative to the two-phonon density of states, the intrinsic linewidth of the LVM in single-isotope  $^{30}\text{Si}$  is actually larger than in natural silicon.

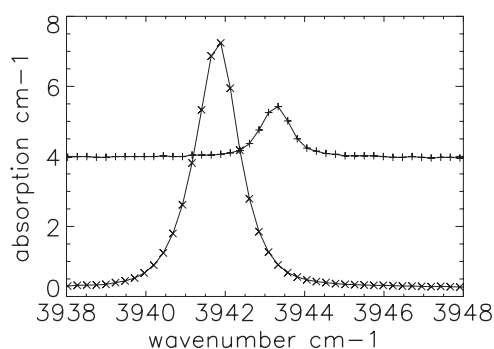
#### 4. Zero-phonon lines

The energy  $E$  of a zero-phonon line (ZPL) is perturbed by strains in the crystal typically at the rate  $\Delta E \sim 10^4\text{ cm}^{-1}$  per unit strain. Residual strains in the crystal are therefore usually expected to produce the linewidth, but, as noted in section 1, the luminescence from excitons bound to shallow donors and acceptors are significantly broadened by isotope disorder—but note that ‘significant’ here means an increase in linewidth due to disorder of  $\sim 0.04\text{ cm}^{-1}$  at these very sharp transitions (Karaiskaj *et al* 2001).

Figure 4 shows the spectrum of the  $3942\text{ cm}^{-1}$  ZPL produced, it is believed, by a complex involving one carbon atom, one oxygen atom and one vacancy (Davies *et al* 1987a). The isotope shift from natural silicon to  $^{30}\text{Si}$  is by  $+1.5\text{ cm}^{-1}$ . Excluding internal transitions at the shallow donors and acceptors (Karaiskaj *et al* 2003b), this is the smallest isotope shift by a factor of five observed to date for a ZPL in silicon (Hayama *et al* 2004a, 2004b, Davies *et al* 2005).

To understand the small size of the shift we note that the shift is partly produced by the volume change in the crystal when the isotopes are changed. This change is by  $\Delta V/V = -1.68 \times 10^{-4}$  (Sozontov *et al* 2001, Wille *et al* 2002), giving a change in energy of the  $3942\text{ cm}^{-1}$  ZPL by  $+1\text{ cm}^{-1}$ , using the deformation potential for the line measured by Davies *et al* (1987b). The remainder of the change arises from the change in the electronic distribution at an optical centre when an electron is excited. This effect may be linked to the temperature dependence of the energy of the ZPL, and when the temperature dependence was reported, it was calculated that the contribution to the isotope shift would be by  $+2\text{ cm}^{-1}$  (Davies *et al* 1987b). The effect is simplified at the  $3942\text{ cm}^{-1}$  band by the absence of any LVMs. The total predicted shift is therefore  $\sim +3\text{ cm}^{-1}$ , very significantly smaller than the  $+7$  to  $+14\text{ cm}^{-1}$  typical in silicon, and in that sense is in good agreement with experiment.

We do not know the molecular structure of the  $3942\text{ cm}^{-1}$  centre. However, the high oscillator strength of the transition (Davies *et al* 1987a), and the lack of any detected higher



**Figure 4.** Absorption in the zero-phonon line of the  $3942\text{ cm}^{-1}$  band. Crosses ( $\times$ ) show the measured absorption in a natural sample, plus signs ( $+$ ) are for  $^{30}\text{Si}$ , displaced vertically by  $4\text{ cm}^{-1}$  for clarity. The fit to the  $^{30}\text{Si}$  data is for a lineshape of the form given in section 4. The fit to the natural sample uses the 15 such curves, weighted according to the random populations by the isotopes of four equivalent sites.

energy absorption lines, suggest that it is a transition between compact electronic states of the centre, rather than the usual pseudo-donor or pseudo-acceptor system familiar in silicon. To estimate the isotope disorder effects we will assume that four neighbours are particularly important. The line on figure 4 through the datum points for the  $^{30}\text{Si}$  spectrum is a lineshape of the form  $I(\nu) = 1/[c + (\nu - \nu_0)^2]^2$ , centred on  $\nu_0$ ; this function frequently fits zero-phonon lineshapes. The full width at half height used for the fit to the  $^{30}\text{Si}$  data is  $0.9\text{ cm}^{-1}$ . In the case of the natural sample, the line through the data is a sum over the 15 possible combinations of the three isotopes on the assumed four isotope sites, using the same lineshape function. The width of each component is  $1.2\text{ cm}^{-1}$ , most probably reflecting slightly greater background strains in the sample. In this model, the isotope disorder in natural silicon broadens the line by  $0.09\text{ cm}^{-1}$ , of the order of 10% of the linewidth after correction for the spectral resolution. In a 50:50  $^{28}\text{Si}$ ,  $^{30}\text{Si}$  crystal, the linewidth in this model would be increased by  $0.4\text{ cm}^{-1}$  compared to a single-isotope sample, which would then be a significant broadening. We note that increasing the number of atoms involved will decrease the range of the disorder and so reduce the line broadening.

The isotope disorder effect is therefore unlikely to be important unless the width of the ZPL is significantly less than  $1\text{ cm}^{-1}$ .

## 5. Summary

We have shown that isotope disorder effects can be detected at the local vibrational mode of substitutional carbon, where the isotope shift between  $^{28}\text{Si}$  and  $^{30}\text{Si}$  is small enough, at  $-3.5\text{ cm}^{-1}$ , to produce a broadening rather than discrete structure. For this LVM, the linewidth is greater in single isotope  $^{30}\text{Si}$  than in the disordered natural silicon. We have suggested that the effect is at least partly due to the increased two-phonon joint density of states function at the LVM value in  $^{30}\text{Si}$ . In the case of zero-phonon lines, we have discussed the  $3942\text{ cm}^{-1}$  line which occurs at a centre that probably has compact electronic states. However, even in this favourable case, the isotope disorder contributes only an estimated  $0.09\text{ cm}^{-1}$  to the linewidth. This contribution is negligible compared to typical strain-broadening effects, but of course it provides an in-principle limit to the widths of the lines in high-purity natural silicon.

It would be desirable to investigate these effects in a 50:50  $^{28}\text{Si}$ ,  $^{30}\text{Si}$  sample.

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