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Phonons, Lattice Instabilities and Superconductivity

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Abstract

The relationship of phonon anomalies and lattice instabilities to high T_c superconductivity is explored. Inelastic neutron scattering studies have revealed an intimate and dramatic relation between certain phonons and lattice transformations, real or incipient, for moderate to high T_c superconductors, which are minimal or nonexistent in very low T_c or nonsuperconductors. Experimental evidence is presented for a wide variety of superconducting materials and possible theoretical explanations for their behaviour are discussed.

Introduction

The electron–phonon interaction is still the most generally accepted mechanism for the occurrence of superconductivity; certainly for the majority of elements, alloys and compounds under investigation today and, in particular, those discussed in this article. It is the basis of the BCS theory (Bardeen *et al.* 1957) which, in turn, stimulated much interest in the vibrational properties of superconductors. The BCS expression for the superconducting transition temperature T_c is given by

$$T_c = 1{\cdot}14\,\Theta \exp(-1/N(0)\,V),$$

where Θ is the characteristic Debye temperature, $N(0)$ is the electronic density of states at the Fermi level and V is the electron–phonon interaction parameter. Since Θ is of the order of a few hundred kelvin, it is obvious that T_c must be very sensitive to the electronic and vibrational properties of the material.

The importance of the phonon spectrum in high T_c superconductivity was emphasized by McMillan (1968) in his classic paper on strong coupling superconductivity in which he explicitly stated the relationship between T_c and the electronic and vibrational properties through λ, the electron–phonon coupling parameter:

$$T_c = \frac{\Theta}{1{\cdot}45} \exp\left(-\frac{1{\cdot}04(1+\lambda)}{\lambda-\mu^*(1+0{\cdot}62\,\lambda)}\right),$$

where μ^* is the Coulomb pseudopotential and λ is defined by

$$\lambda = 2\int_0^{\omega_0} \alpha^2(\omega)\,F(\omega)\,\omega^{-1}\,\mathrm{d}\omega.$$

Here $\alpha^2(\omega)F(\omega)$ is the electron–phonon spectral function and is obtainable from electron tunnelling experiments on superconductors; $F(\omega)$ is the phonon density of

states and is obtainable from inelastic neutron scattering experiments; ω_0 is the maximum phonon frequency in the crystal. There are often experimental difficulties in obtaining either or both of α^2F and F.

With certain assumptions and suitable approximations McMillan (1968) showed that

$$\lambda = N(0)\langle I^2\rangle / M\langle\omega^2\rangle ,$$

where $\langle I^2\rangle$ is the electron–phonon matrix element averaged over the Fermi surface, M is the ionic mass and

$$\langle\omega^2\rangle \equiv \int \alpha^2(\omega)\, F(\omega)\, \omega \, \mathrm{d}\omega \Big/ \int \alpha^2(\omega)\, F(\omega)\, \omega^{-1} \, \mathrm{d}\omega$$

$$\approx \int F(\omega)\, \omega \, \mathrm{d}\omega \Big/ \int F(\omega)\, \omega^{-1} \, \mathrm{d}\omega .$$

From an analysis of extensive data on transition metal alloys McMillan concluded that $N(0)\langle I^2\rangle$ was essentially a constant and, therefore, that the variations in λ and hence T_c were primarily due to 'certain' phonon frequencies, but it was not clear which phonon frequencies were the relevant ones. It was about this time that inelastic neutron scattering results (Ng and Brockhouse 1968; Powell *et al.* 1968; Smith and Gläser 1970) showed that there were, indeed, regions in the Brillouin zone where the phonon frequencies were anomalously 'soft' in moderate to high T_c superconductors but were 'normal' in low T_c or nonsuperconductors. The higher the T_c the larger was the phonon softening, resulting ultimately in a lattice instability—perhaps the cause of the lattice transformations in the very high T_c A-15 compounds V_3Si and Nb_3Sn (Matthias *et al.* 1971; Phillips 1971; Testardi 1972; Smith *et al.* 1976*b*; Smith and Finlayson 1980). The idea that the attainment of high T_c superconductivity may, in fact, be limited by lattice instabilities has been proposed for some time. Ultrasonic measurements, specific heat measurements and thermal expansion data, along with crystallographic studies, have provided some of the first experimental evidence in this regard, aside from the difficulty of materials preparation in the first place.

Weber (1973*a*) questioned the assumption that $N(0)\langle I^2\rangle$ was a constant for the Nb–Mo system and for the transition metal carbides, and further analysis of the McMillan (1968) equation by Allen and Dynes (1975) verified that $N(0)\langle I^2\rangle$ is not a constant and is probably more responsible for variations in T_c than is $M\langle\omega^2\rangle$. McMillan himself, in his article, had emphasized that the phonon frequencies are extremely sensitive to small changes in the electronic properties. Apparently, the phonon anomalies are more an effect than a cause of superconductivity but, as we shall see in the following sections, a relation between large phonon anomalies and high T_c definitely exists and if one can understand the source of the anomalies it should lead to a better understanding of superconductivity. Progress in the theoretical interpretation of these results is, indeed, being made and will be discussed below.

As mentioned above, there are often difficulties in obtaining $\alpha^2F(\omega)$ from superconducting tunnelling or $F(\omega)$ from neutron data even if the dispersion curves have been measured. Allen (1972) has shown that λ can be directly and simply related to the electron–phonon contribution γ_i^{ep} to the phonon linewidths γ_i and phonon frequencies ω_i by

$$\lambda = \sum_i \gamma_i^{ep}(\boldsymbol{q}) / N(0)\, \pi\, \omega_i^2(\boldsymbol{q}) .$$

It would be a horrendous task to determine λ in this way even if other contributions to the linewidth were negligible. Nevertheless, it has been possible to confirm this concept by comparing a selected number of experimental measurements with the theoretical predictions (Butler *et al.* 1977) based on a detailed knowledge of the band structure and Fermi surface.

Experimental Method

Thermal neutrons are particularly well suited for studying the lattice vibrations in solids, for their energies are of the order of the phonon energies and their wavelengths are of the order of the lattice spacings. The changes on scattering are large and relatively easy to measure under conditions of one-phonon scattering (Dolling 1974), in contrast to X-ray thermal diffuse scattering.

The conditions for conservation of crystal momentum and energy require that

$$\boldsymbol{Q} = \boldsymbol{k}_0 - \boldsymbol{k}' = 2\pi\boldsymbol{\tau} + \boldsymbol{q}, \qquad \Delta E = h\nu(\boldsymbol{q}) = h^2(k_0^2 - k'^2)/2m,$$

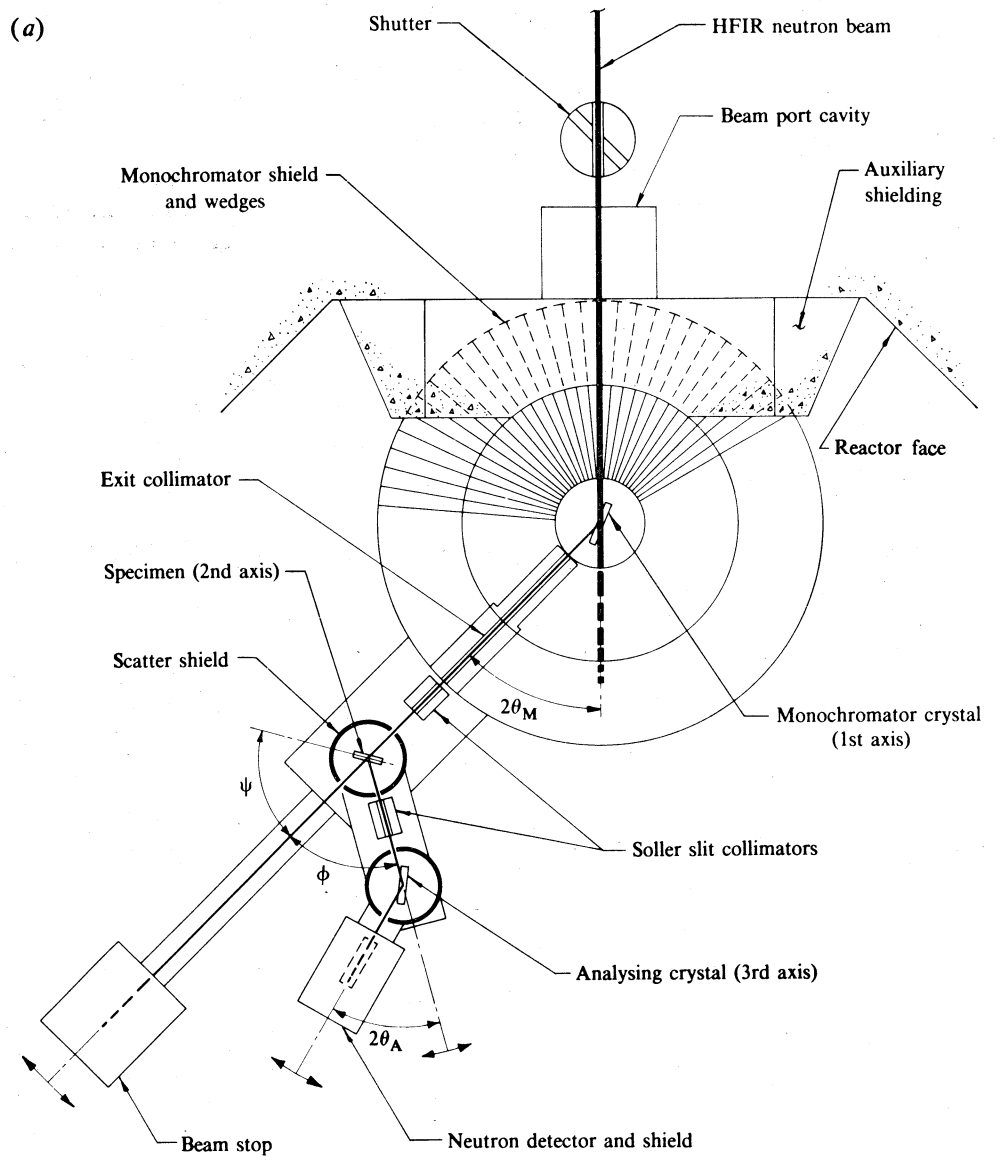
where $\boldsymbol{k}_0$ is the momentum of the incoming neutron, $\boldsymbol{k}'$ is the momentum of the scattered neutron, $h\nu$ is the energy of the phonon with wave vector $\boldsymbol{q}$, and $\boldsymbol{\tau}$ is a reciprocal lattice vector. These quantities are directly and simply related to the experimentally accessible angle variables shown in Fig. 1*a*. The dispersion relation is $\nu_j(\boldsymbol{q})$ where j denotes the phonon branch for a given direction in the crystal. The mapping out of the phonon branches is indicated by the vector diagram in Fig. 1*b* (related to the Ewald sphere construction) and a schematic ν versus q plot (Fig. 1*c*). When the ellipsoid (representing the instrumental resolution function) intersects a given phonon branch a peak occurs in the neutron scattering intensity (Fig. 1*d*). The slope $\Delta\nu/\Delta q$ of the straight line intersecting the origin is the velocity of sound as $q \to 0$.

Superconducting Materials

Transition Metal Carbides

Historically, the first phonon spectra of a moderately high T_c superconductor were measured on f.c.c. Pb (Brockhouse *et al.* 1962) and then b.c.c. Nb (Nakagawa and Woods 1963). It was noted that the dispersion curves were more complex than those for other simpler materials, such as f.c.c. Al and b.c.c. Na, but there were no suitable theoretical models other than the Born–von Kárman models with many parameters. The discovery of phonon anomalies in the superconducting transition metal carbides (Smith and Gläser 1970; Smith 1972) led to a successful phenomenological model not only for the carbides (Weber *et al.* 1972; Weber 1973*b*) but for the Nb–Mo alloys as well (Weber 1973*a*). This stimulated much theoretical work in the last decade on the microscopic formalism of phonon spectra in superconductors.

The transition metal carbides are an interesting class of materials. Some of them are moderate to high T_c superconductors (e.g. NbC and TaC with $T_c \approx 10$ K), as are the nitrides (T_c of $NbC_xN_{1-x} = 17$ K for $x = 0{\cdot}30$). They have bonding properties typical of metallic, covalent and ionic materials simultaneously. Their melting points are some of the highest known (T_m of TaC = 4256 K). They have the f.c.c. NaCl structure, so it is, perhaps, instructive to see what the dispersion curves look like for a very simple substance, namely LiF (see Fig. 2).



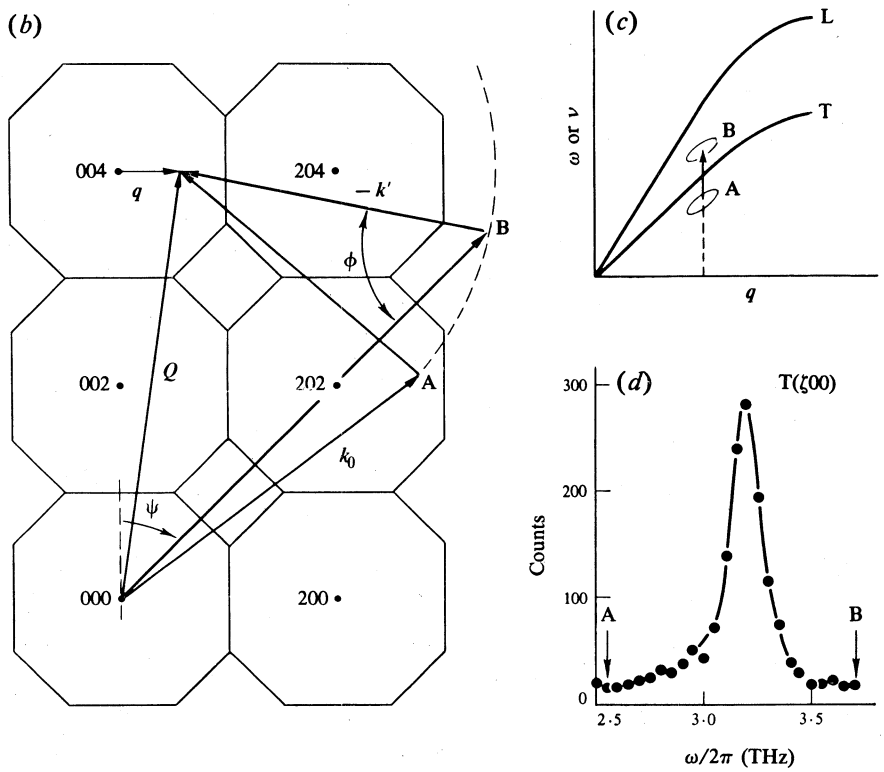
Figs 1*a*–1*d*. Schematic diagrams of:

(*a*) a HB-3 triple axis spectrometer;

(*b*) a vector mapping of a typical phonon scan;

(*c*) a plot of frequency ν versus wave vector q;

(*d*) a peak in the neutron scattering intensity.

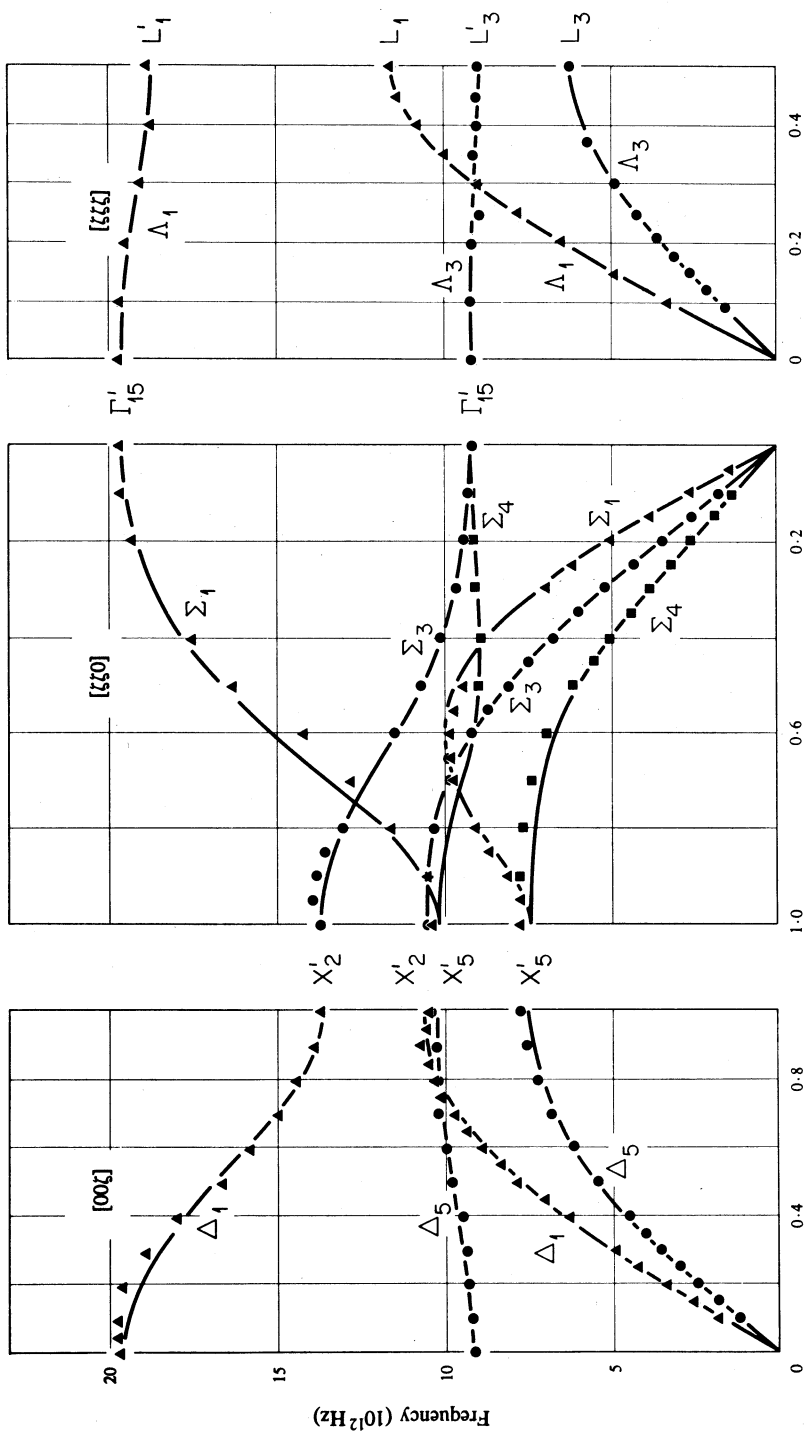


Figs 1*b*–1*d*

The compound LiF is the simplest of the alkali halides and can be described by the 'simple' shell model (Dolling *et al.* 1968). It is a parametrized model but has a firm microscopic basis. The slopes of the acoustic modes agree very well with the velocity of sound measurements, as do the frequencies of the longitudinal optic (LO) and transverse optic (TO) modes when compared with the infrared absorption and reflectivity measurements. The large splitting of the LO and TO modes at $q = 0$ is due to the well-known Lyddane–Sachs–Teller splitting in the long-wavelength or $q = 0$ limit and is indicative of the large macroscopic field in the ionic crystal.

A similar model with long-wavelength conduction electron screening was used to calculate the dispersion curves of ZrC (Fig. 3*a*), a nonsuperconducting metal (Smith *et al.* 1976*a*). In this case there is no macroscopic field in the crystal, for the ions are screened by the conduction electrons; therefore, there is no splitting of the LO and TO frequencies and the modes are degenerate at $q = 0$, as required by symmetry. The phonon dispersion curves for superconducting NbC are shown in Fig. 3*b*. Notice the anomalous dips in the LA(001) branch at $\zeta = 0{\cdot}6$, in the LA(110) branch at $\zeta = 0{\cdot}6$ and in both the LA(111) and TA(111) branches at $\zeta = 0{\cdot}5$. The dips are reduced in size for nonstoichiometric NbC_x (T_c decreases) and are absent when $x = 0{\cdot}75$ ($T_c = 0$).

An explanation of the anomalous dips in the superconducting carbides in terms of a phenomenological 'double-shell' model was put forward by Weber *et al.* (1972). The extra shells (or 'supershells') on the transition metal atoms interact attractively



Reduced wave vector coordinate ζ

Fig. 2. Phonon dispersion curves for LiF at 298 K. The reduced wave vector coordinate $\zeta = a_0 q/2\pi$.

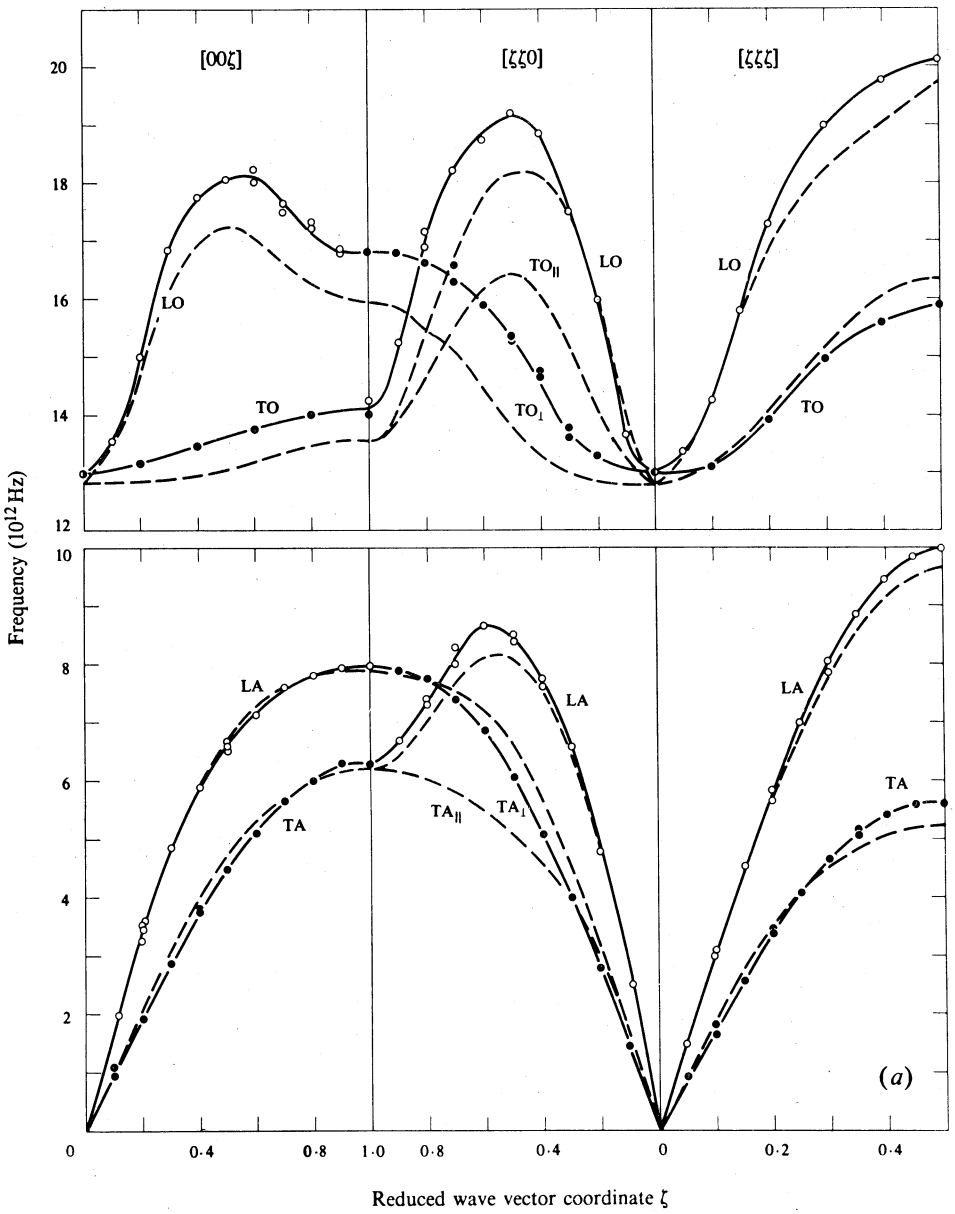
with neighbouring supershells, including second neighbours on the metal sublattice, producing a resonance-like behaviour in the dynamical matrix for the above mentioned values of ζ. The excellent agreement with the experimental data for TaC is shown in Fig. 3*c*. This model is rather complicated with many adjustable parameters, but does have the advantage of identifying the components responsible for the phonon anomalies.

Transition Metal Alloys

The most promising high T_c superconductors involve the transition metal elements, alloys and compounds. Matthias (1955) showed empirically for a large number of alloys and compounds of the 4d and 5d transition metals and, to a limited extent, the nonmagnetic 3d metals that maximum T_c values occur when the electron per atom ratio (e/a) is about 4·8 and 6·6 with minima near 4, 5·7 and 7·5, where the formal valence is taken to be 4, 5 and 6 for Zr, Nb and Mo respectively, etc. We shall see that the phonon anomalies can also be correlated with this rule. Fig. 4 shows the LA modes in the [001] direction for the fifth and sixth column elements. The anomalous dips near $\zeta = 0{\cdot}7$ for the higher T_c metals V, Nb and Ta are absent in the nonsuperconducting or low T_c metals Cr, Mo and W. In an extensive series of neutron scattering experiments on the Nb–Mo alloys, Powell *et al.* (1968) showed that the magnitude of the dip in Nb decreased with increasing Mo concentration and disappeared completely at $Nb_{0{\cdot}25}Mo_{0{\cdot}75}$ ($T_c = 0$) and perhaps was appearing again in pure Mo ($T_c = 0{\cdot}9$ K) at the zone boundary point H (Fig. 5). Upon alloying Mo with Re, Smith *et al.* (1976*a*) showed that the H point was indeed anomalous, dramatically softening to almost half its value for $Mo_{0{\cdot}85}Re_{0{\cdot}15}$ and decreasing further for $Mo_{0{\cdot}75}Re_{0{\cdot}25}$ (with T_c increasing to ≈ 10 K). For greater concentrations, T_c increases but the system is no longer single phase, indicating an instability in the b.c.c. lattice. Traylor *et al.* (1975) showed that similar behaviour exists in the Nb-rich–Zr system. In contrast to this behaviour, the b.c.c. phase is stable all the way across the Nb–Mo system, where the values of T_c are minimal.

Although the h.c.p. structure is not generally considered favourable for high T_c superconductivity, technetium is an exception with $T_c = 8$ K ($e/a = 7$). The phonon dispersion curves have been measured (Smith *et al.* 1974) and a pronounced softening was observed which is of the long-wavelength optic mode in the c direction; an out of phase motion between neighbouring hexagonal planes. A comparison (Smith *et al.* 1976*a*) of this branch with that of Ru ($e/a = 8$), a very low T_c superconductor (Fig. 6), shows a striking difference in their behaviour and reiterates the lattice instability concept and its correlation with the e/a ratio propounded by Matthias (1955). It would be very desirable to study single crystals of Tc alloyed with neighbouring elements, for small amounts of Mo and Nb increase T_c dramatically, whereas, alloying Tc with elements to the right of it in the periodic table lowers the T_c value.

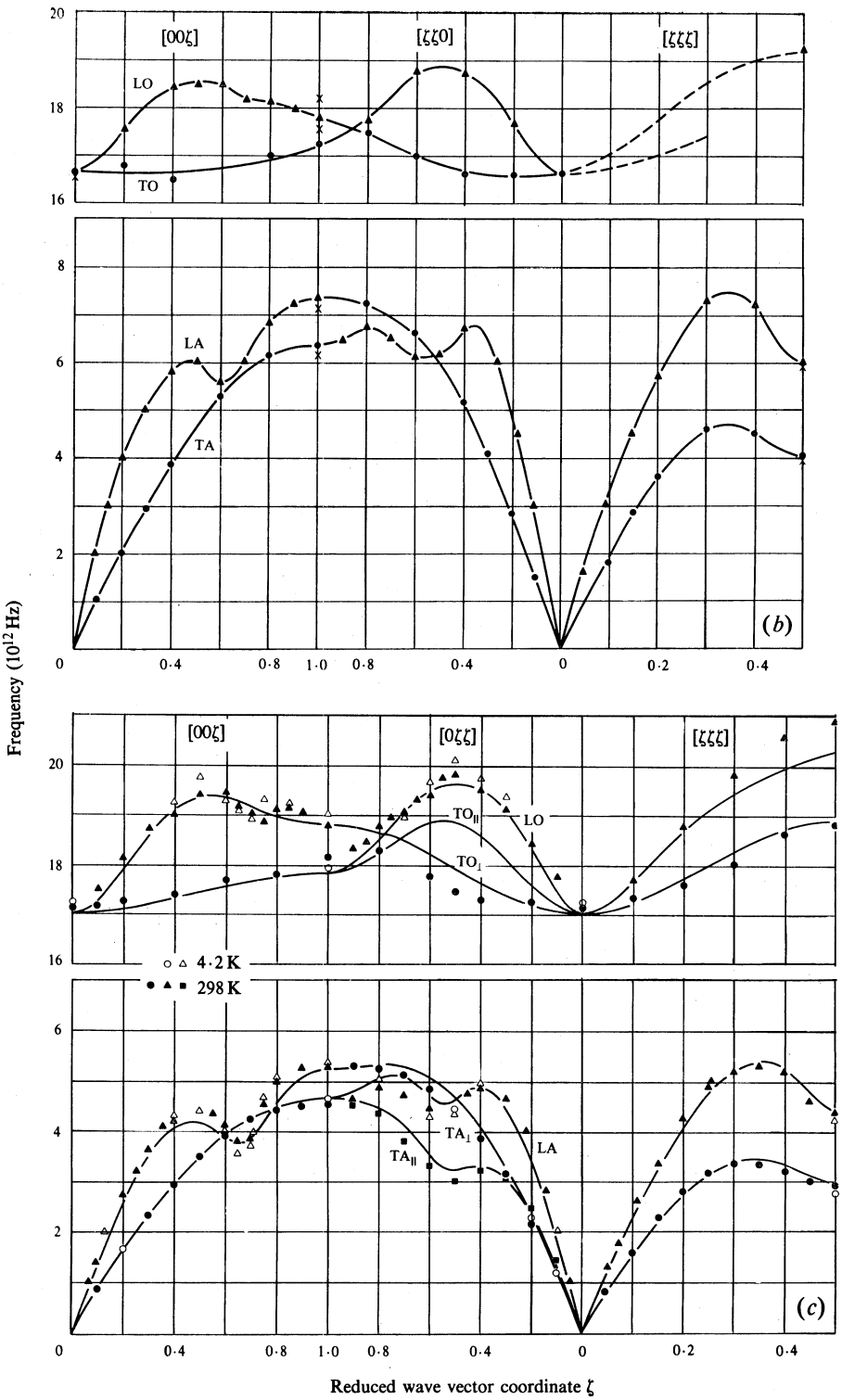
A recent study of the temperature dependence of the above mode to 10 K revealed a softening from 3·8 to 2·4 THz. This softening is much greater than that observed for other superconductors with the exception of V_3Si (Shirane and Axe 1971) and Nb_3Sn (Axe and Shirane 1973); however, little is known about the detailed phonon behaviour in the A-15 system, although such studies are currently underway.



Figs 3*a*–3*c*. Phonon dispersion curves for (*a*) ZrC at room temperature, (*b*) NbC at room temperature and (*c*) TaC at 4·2 and 298 K. The dashed and solid curves in (*a*) and (*c*) respectively are calculations based on the double-shell model of Weber *et al.* (1972).

α-uranium

A puzzle concerning the stability of the α-U lattice has recently been resolved by neutron scattering studies. Ultrasonic, heat capacity and thermal expansion measurements on single crystals suggested some type of phase transition but X-ray and previous neutron diffraction studies did not indicate any crystal structure changes,



Figs 3*b* and 3*c*

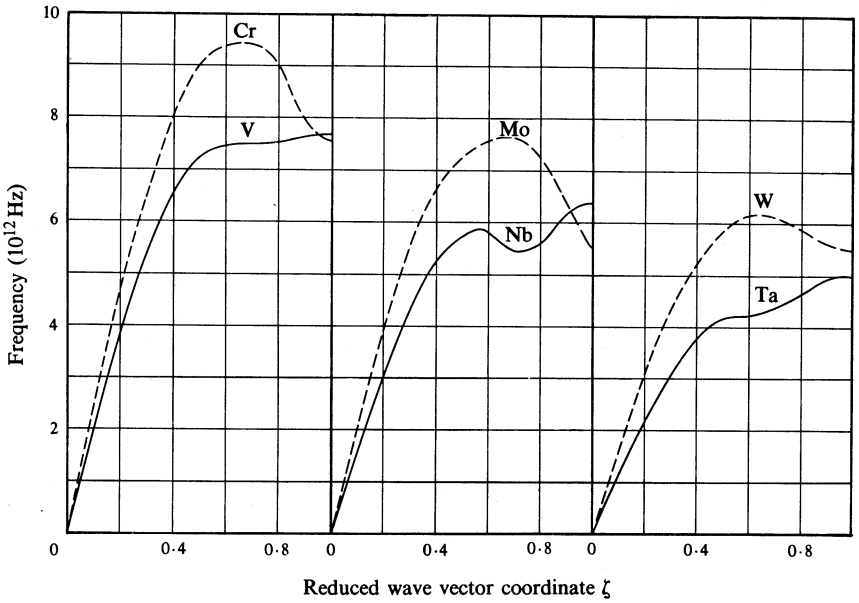


Fig. 4. Longitudinal acoustic modes in the [001] direction for Group V and Group VI transition metals.

although a net increase in volume at low temperatures was noted. Lattice dynamical studies by Crummett *et al.* (1979), utilising inelastic neutron scattering techniques, revealed a large anomalous dip in the LO[100] optic modes. By itself this observation would indicate a possible contradiction to the correlation of high T_c materials with phonon anomalies, for single crystals of α-U exhibit very low T_c superconductivity. However, application of modest pressures increases T_c to over 2 K (Smith and Fisher 1973) and also reduces the temperature of the ultrasonic anomalies (Fisher and Dever 1970). This was taken to mean that another phase or phases coexisting with α-U were being suppressed by pressure and that α-U itself was exhibiting superconductivity. A neutron scattering study of the temperature dependence of the anomalous dip of the LO optic modes showed (Fig. 7) a softening and eventually a condensation into a charge density wave (CDW) at low temperatures (Smith *et al.* 1980). It is now believed that this CDW is suppressed by pressure and a further neutron diffraction study of α-U under pressure is currently underway. Although the phonon anomaly is large and the T_c is rather modest, there are other factors which affect the electron–phonon coupling parameter λ; for α-U, perhaps, it is a low electronic density of states at the Fermi level. Clearly, there is a need for more theoretical and experimental work on uranium.

Non-Transition Metals

Although the above discussions have been concerned with transition metals and α-U, an actinide, similar lattice instabilities or phonon anomalies have been observed in non-transition metals such as Pb, In, Sn and Hg and in some of their alloys. The most notable example is the Pb–Tl system (Ng and Brockhouse 1968), where alloying Pb with Tl decreases T_c and reduces the size of the phonon anomalies (Fig. 8). This system is very stable, exhibiting a solid solution over most of the composition range

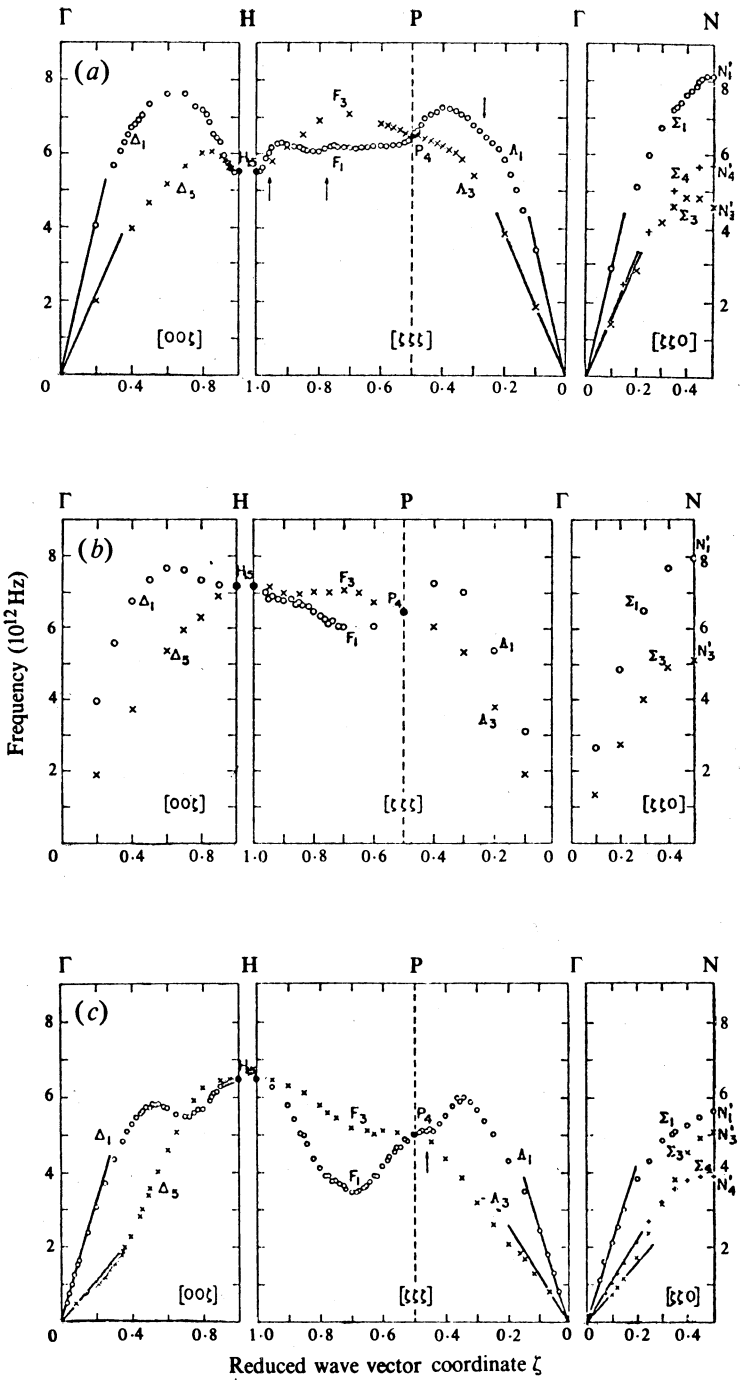


Fig. 5. Phonon dispersion curves for (*a*) Mo, (*b*) $Nb_{0.25}Mo_{0.75}$ and (*c*) Nb at room temperature. The plotted crosses (circles) represent transverse (longitudinal) phonons.

in the f.c.c. structure. However, upon alloying Pb with Bi, the T_c increases, the phonon anomalies deepen and the f.c.c. phase becomes unstable. Lattice dynamical calculations by Vosko *et al.* (1965) suggested that the anomalous regions in the Pb dispersion curves are due to negative contributions of the electron–ion interactions to the dynamical matrix.

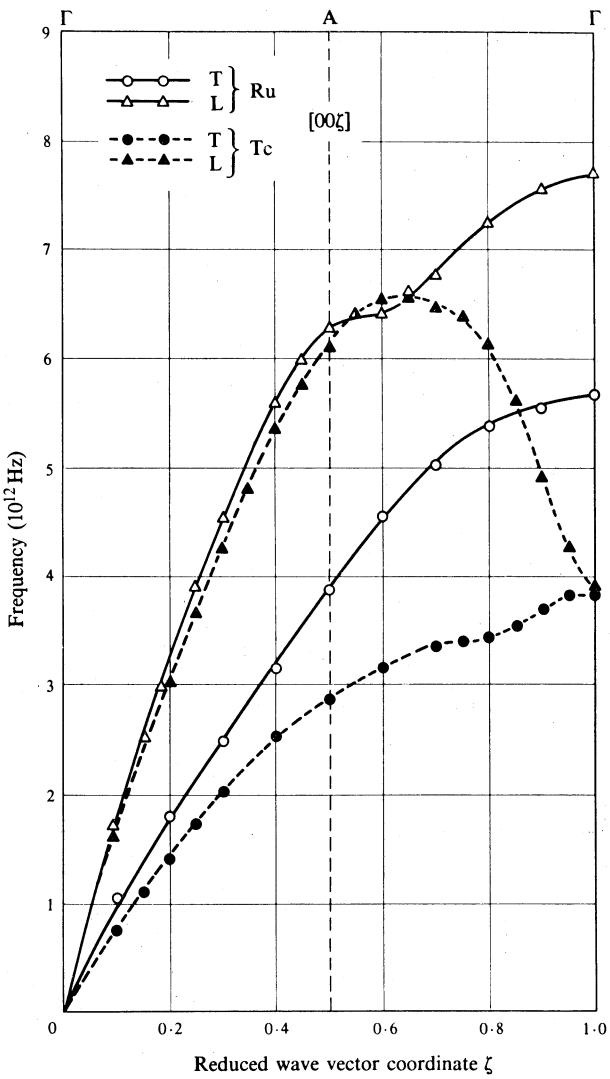


Fig. 6. Dispersion curves in the *c* direction for Tc and Ru at room temperature. The plotted circles (triangles) represent transverse (longitudinal) phonons.

Theory

There is no doubt that a better knowledge of the electron–phonon interaction in metals would be very beneficial to understanding many solid state properties in normal metals as well as superconducting metals. These properties are ultimately

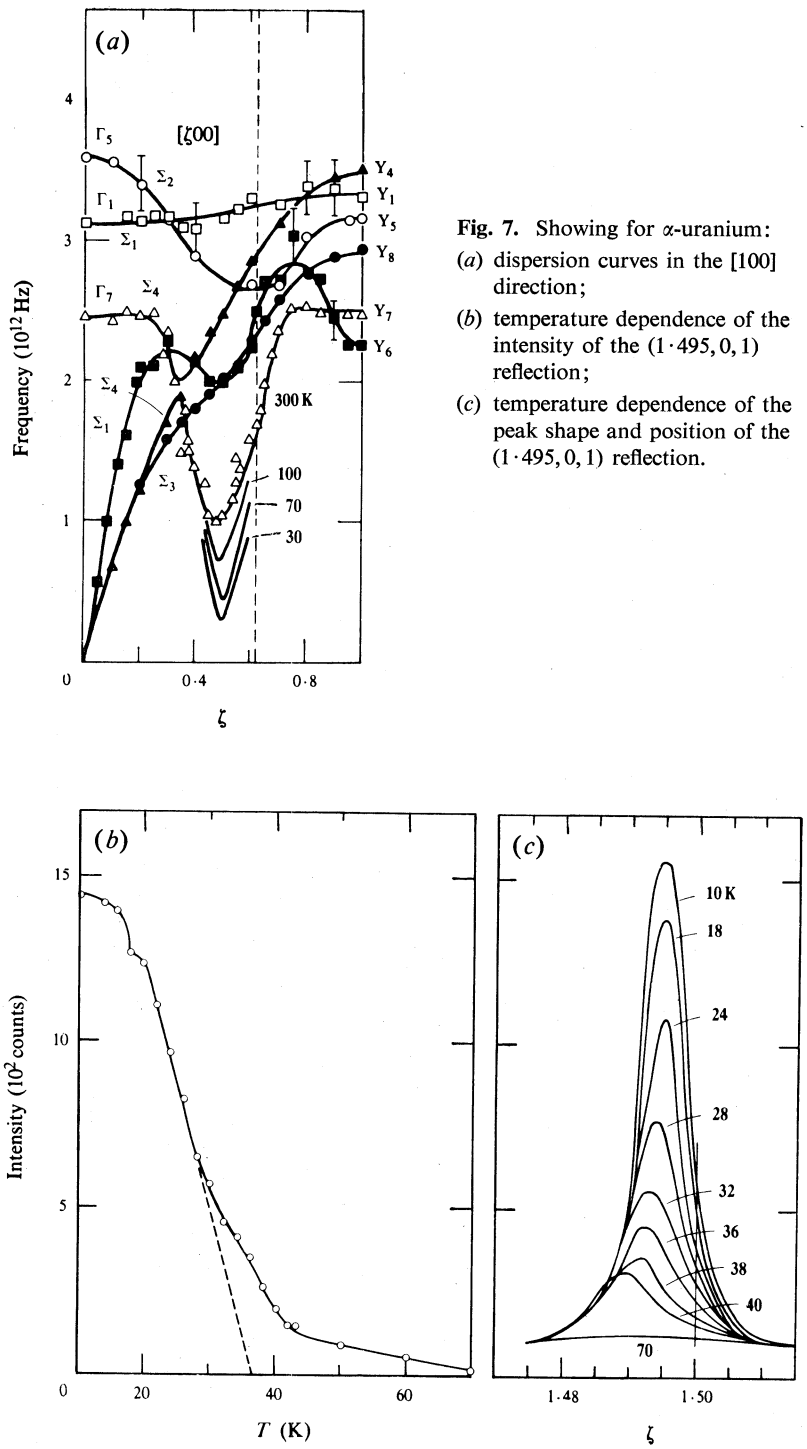


Fig. 7. Showing for α-uranium:
(*a*) dispersion curves in the [100] direction;
(*b*) temperature dependence of the intensity of the (1·495, 0, 1) reflection;
(*c*) temperature dependence of the peak shape and position of the (1·495, 0, 1) reflection.

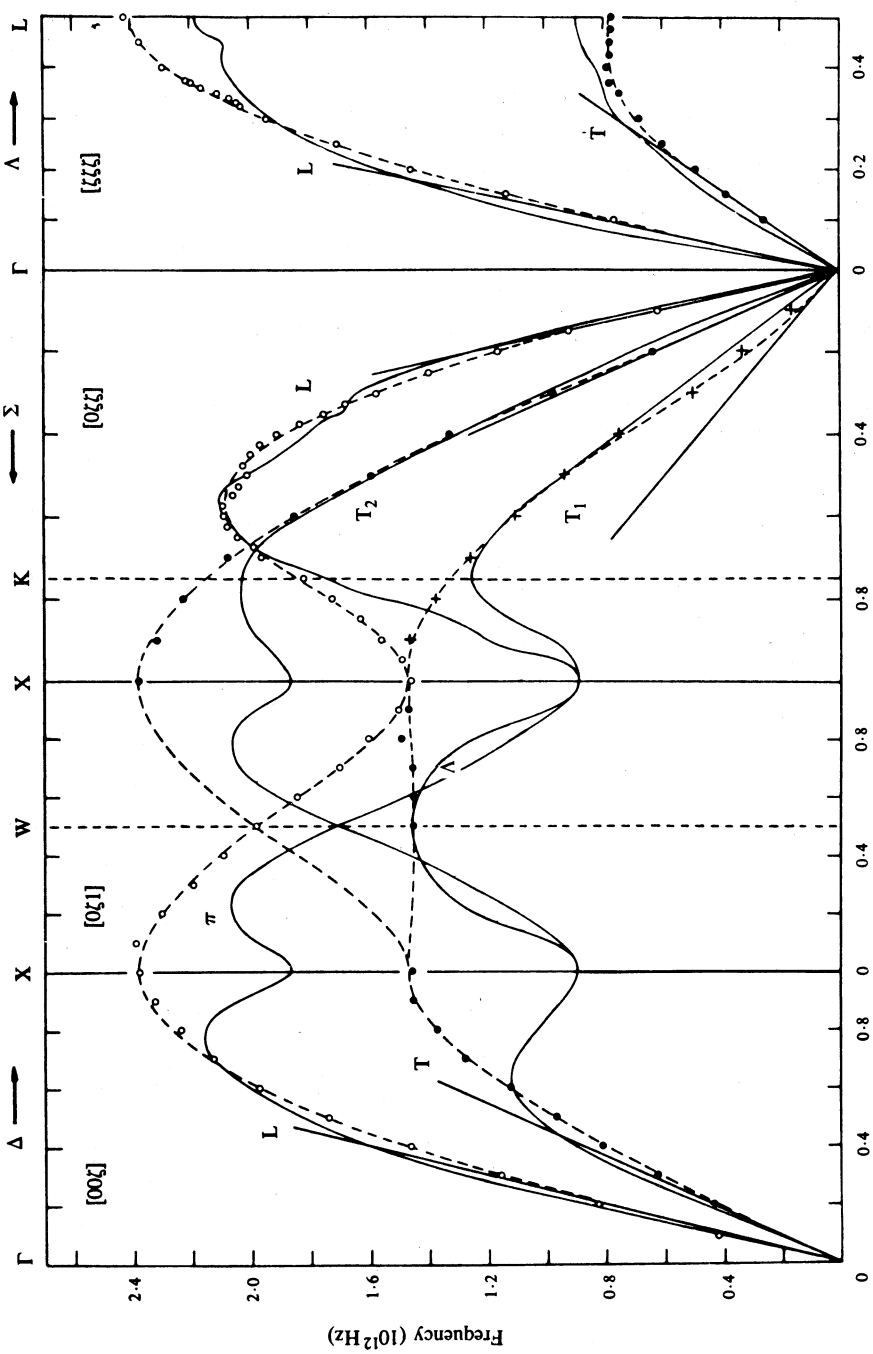


Fig. 8. Comparison of phonon dispersion curves at 100 K for Pb (solid curves) and $Pb_{0\cdot4}Tl_{0\cdot6}$ alloys (points) (from Ng and Brockhouse 1968).

related to the detailed electronic structures of the elements, their alloys and compounds, as are their observed crystal structures and phonon spectra. Major advances have been made in calculating electronic structures in recent years and serious attempts are being made to calculate the electron–phonon interaction from a first-principles microscopic basis (Butler 1977; Butler *et al.* 1979; Allen 1980; Sinha 1980). Some may feel only the details are left to be filled in, but it is precisely these details which are responsible for many of the quantities of interest, for example, phonon dispersion curves.

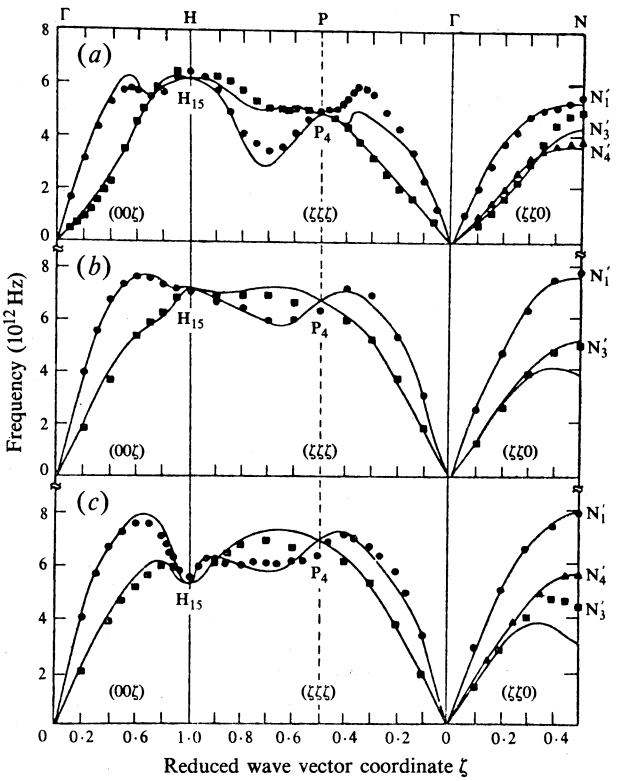


Fig. 9. Phonon dispersion curves at room temperature for (*a*) Nb, (*b*) $Nb_{0.25}Mo_{0.75}$ and (*c*) Mo. The calculated results of Varma and Weber (1977) are compared here with experimental data from Powell *et al.* (1968).

The expressions for the frequencies of the normal modes of vibration in a crystal, i.e. the dynamical matrix, are intimately related to the type and degree of bonding, to electronic and ionic polarizabilities, as well as to screening of the ion–electron and electron–electron interactions. However, these quantities themselves are changing during the course of the lattice vibrations and must have an effect on the electronic band structures, which in turn affect the phonon spectra. Here also, serious attempts are being made to calculate the details of the phonon spectra from first principles, but the job is a horrendous one. However, one can use the experimentally determined phonon dispersion curves as a guide in deciding which electronic properties predominantly affect the phonon spectra and ultimately contribute to the electron–phonon interaction.

Varma and Weber (1977, 1979) have presented a new formulation of the lattice dynamics of transition metal alloys and compounds, based on a nonorthogonal tight binding representation of the d electrons. They separate the contributions to the dynamical matrix into strong short range terms, which are parametrized, and long range terms, attributed to overlap forces of the 'valence' electrons, which are calculated from first principles from a knowledge of the electronic wavefunctions. Fig. 9 shows the impressive results for Nb, Mo and $Nb_{0.25}Mo_{0.75}$. All the important features in the experimental curves are seen to be contained in the calculations. From

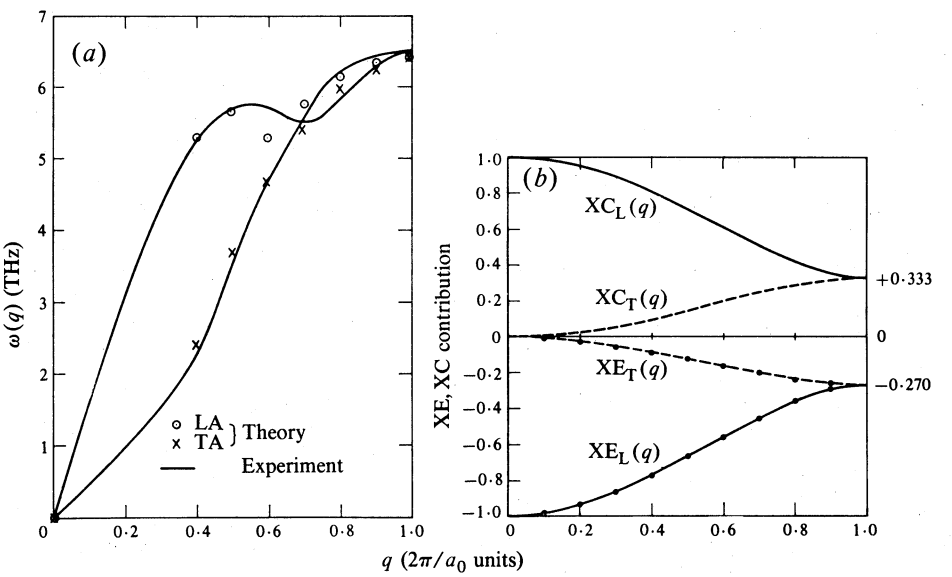


Fig. 10. Showing for Nb along the [100] direction: (*a*) a comparison between theory and experiment for the longitudinal and transverse acoustic modes, and (*b*) the contributions to the phonon frequency from the ion–ion (XC) and electron–ion (XE) interactions; the subscripts L and T refer to the longitudinal and transverse modes respectively (from Cooke 1978).

a detailed analysis of their calculations they conclude that the phonon anomalies are not merely *large* Kohn anomalies attributable to purely Fermi surface effects (i.e. Fermi surface 'nesting'), but are due to band structure effects near the Fermi surface—and the flatter the bands the larger the anomalies.

Cooke (1978*a*, 1978*b*) has made a parameterless first-principles calculation of the phonon dispersion curves of Nb in the [100] direction. It is based on the RPA dielectric response formalism which generates a natural decomposition of the dynamical matrix into two parts, one due to direct ion–ion interactions (XC) and the other due to the electron–ion interactions (XE). The ion–ion interactions are relatively straightforward to calculate, but the electron–ion interactions are more difficult to determine, requiring the electronic energies, wavefunctions and matrix elements based on realistic electron–ion and crystal potentials. The comparisons between theory and experiment for modes in the [100] direction are shown in Fig. 10*a*. The agreement here is also very impressive, particularly so, considering there are no adjustable parameters. The contributions of XC and XE as a function of q for the longitudinal and transverse modes are shown in Fig. 10*b*. Notice that there are

no anomalies or dips in these curves, yet a dip is produced in the LA modes as a result of XC_L+XE_L. The overall phonon bandwidth is reproduced exactly. The calculations in the [110] and [111] directions are far more difficult because of convergence problems.

Cooke (1980) has recently made similar calculations for Mo, and the results are shown in Fig. 11. A potential based on the local-density approximation to exchange and correlation was used in the calculations. As in Nb, the energy bands and wavefunctions were generated from a KKR band structure formalism. A rigid band

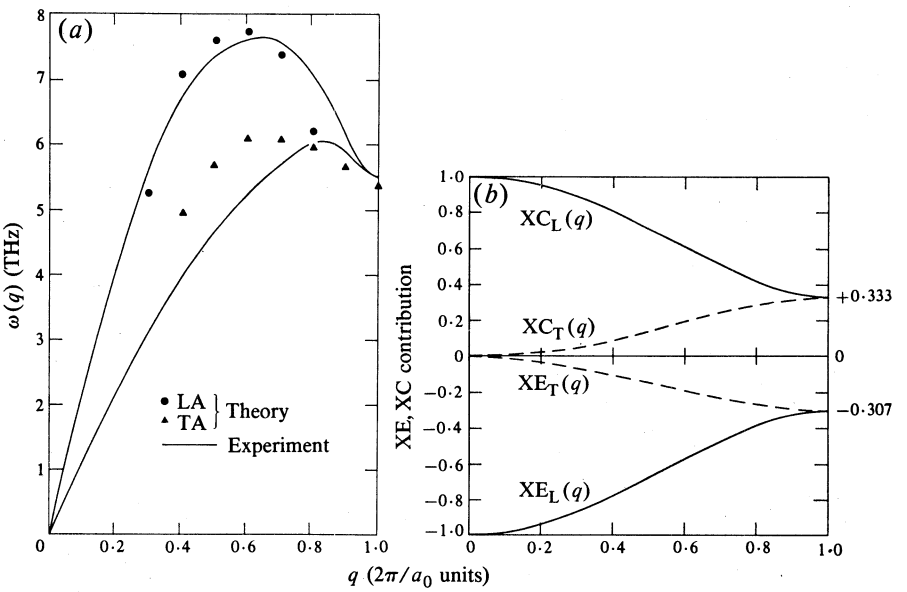


Fig. 11. Showing for Mo along the [100] direction: (*a*) a comparison between theory and experiment for the longitudinal and transverse acoustic modes, and (*b*) the contributions to the phonon frequency from the ion–ion (XC) and electron–ion (XE) interactions for the longitudinal (L) and transverse (T) modes (from Cooke 1980).

calculation using the potential employed in the Nb calculations was also made with similar, but not as good, results; the phonon bandwidth was too high compared with the experimental results. In both cases, however, the anomalous dips at the zone boundary point H were reproduced. Although it was shown, as in Nb, that the anomaly is not associated with Fermi surface nesting, the terms responsible for the dip are not revealed.

Concluding Remarks

There seems to be little doubt that a relationship exists between the observed anomalies in phonon dispersion curves and the superconducting transition temperature in superconductors. The anomalies very likely reflect lattice instabilities in high T_c superconductors and are related somehow to the detailed electronic properties of the metal, alloy or compound, particularly the properties relating to chemical bonding.

Inelastic neutron scattering studies have played a dominant role in revealing the anomalous behaviour. These studies in turn have encouraged the theorists to develop new lattice dynamical and microscopic theories to explain the experimental results and, as shown in the previous section, with a good degree of success. Undoubtedly, the microscopic basis of these theories can also be used to predict other interesting properties of the metallic state.

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