Temperature dependence of phonon intensities in tantalum below 4.2 K

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An unexpected temperature dependence of the phonon peak intensity in tantalum, as measured by neutron scattering, is observed between 1.2 and 6 K, that is, in a temperature range where the phonon self-energy is dominated by the electron-phonon interaction. A possible relation between the observed behavior of the phonon intensity and the results of a tunneling experiment is presented. Possible sources of the anomalous behavior presently observed are also considered.

I. INTRODUCTION

The study of lattice dynamics in transition metals is of great importance in order to obtain useful information on the structural stability of the system as well as on its behavior at the superconducting transition[1] when present. Neutron inelastic scattering is a well established tool to determine phonon-dispersion curves and phonon interactions through the analysis of the displacement-displacement correlation function, which is simply related to the neutron cross section.[2] A detailed investigation of the phonon response function over a wide energy range is generally not necessary in studies of phonon lifetime.[3–5] However, inspection of a wide momentum-energy range turns out to be useful in order to analyze the energy dependence of the phonon self-energy,[6–8] and it is particularly relevant when considering the electron-phonon interaction in systems undergoing a transition, like a structural transition or, to a lesser extent, a superconducting transition.

At low temperature, when no structural transitions are present, the lattice dynamics is essentially harmonic, so that the phonon self-energy is determined by the electron-phonon interaction. The average electron-phonon interaction is measured by the electron-phonon coupling parameter λ which can be determined by a proper average of the phonon lifetime.[9] Among superconducting transition metals, tantalum shows a remarkable disagreement in λ as deduced in the superconducting phase from the transition temperature through McMillan's equation and from electronic specific-heat or resistivity measurements,[10] that is, above the superconducting transition. Such a discrepancy could be attributed to an anomalous behavior of the phonon density of states or of the phonon lifetime at low temperature.

It should be also observed that the phonon density of states $F(\omega)$ weighted with the electron-phonon coupling function $\alpha^2(\omega)$, as measured by a tunneling experiment,[11] shows important differences when compared to the density of states derived from a room temperature neutron inelastic scattering experiment.[12] Such a discrepancy could be explained by a strong energy dependence of $\alpha(\omega)$, even though $\alpha(\omega)$ has been found to be almost constant in other transition metals.[9,11] In view of the discussed disagreements found in this system, a low-temperature study of the phonon groups seemed to be worthwhile. Indeed both the anomalous behavior of λ and that of the tunneling density of states could be due to some change of the phonon response function.

Recently it has been shown that the PRISMA (Refs. 13 and 14) spectrometer, installed at the ISIS Spallation Neutron Source (Chilton, U.K.), allows for investigations of extended portions of momentum-energy space simultaneously with good performance characteristics. Thanks to the intrinsic nature of the Progetto dell'Istituto di Struttura della Materia (PRISMA) spectrometer, which is a time-of-flight instrument, it is possible to perform studies of temperature-dependent phenomena maintaining the sample in a fixed position, thus reducing incidental spurious effects introduced by possible instrumental misalignments during different scans. In addition it is possible to have a rather complete overview of the one-phonon response function, thus allowing for an easy identification of the interesting features even if their location in the Brillouin zone is not *a priori* known.

II. EXPERIMENT AND DATA REDUCTION

The inelastic-scattering experiment has been performed using the PRISMA spectrometer, employing 15 independent Ge(111) analyzers[14] and 15 ^{3}He detectors. The sample was a cylinder, 1.2 cm in diameter and 5 cm in height, with the [001] axis vertical. The phonons were measured along the [100] direction around the (420) Bragg peak. The temperature was varied between 1.2 K and room temperature and the stability was within ± 0.1 K. Measurements have been performed at four temperatures, namely 1.2, 4.2, 6 K, and room temperature. Several spectrometer configurations have been employed in order to check the effect of different reciprocal-space cuts.

The data collected at room temperature are reported on the dispersion curve shown in Fig. 1 in comparison with a force-constant fit to the older data of Ref. 12. The agreement between the two measurements is reasonably good, although the transverse branch of the previous data,[12] which consists of four points in the reduced q range 0.2–0.5, is systematically lower than the present

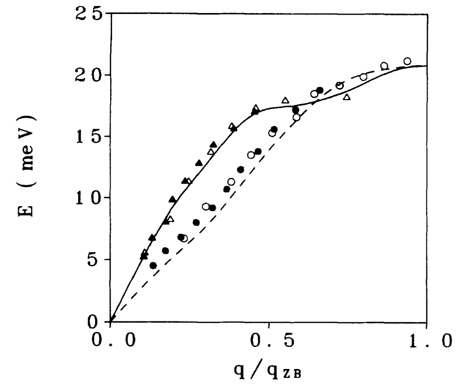

FIG. 1. Room-temperature phonon-dispersion curve for tantalum in the [00ζ] direction. Present transverse-acoustic-phonon (TA) energies: energy loss (circles) and energy gain (dots). Present longitudinal-acoustic-phonon (LA) energies: energy loss (triangles), energy gain (full triangles). Transverse-phonon energies from Ref. 12: dashed line fitted to the data. Longitudinal phonon energies from Ref. 12: continuous line fitted to the data.

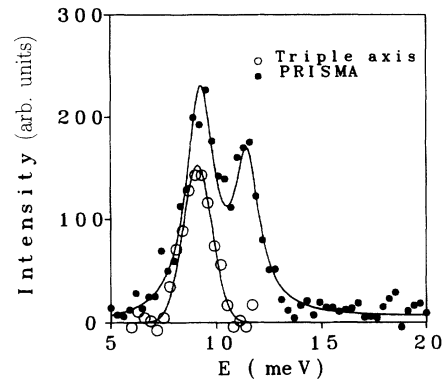

FIG. 2. Room-temperature phonon groups along the [00ζ] direction at $\zeta=0.3$ for Ta, measured by conventional triple-axis and PRISMA spectrometer. The PRISMA scan is such that the reduced wave vector at the transverse-phonon peak position is $\zeta\simeq0.31$. Full lines are fits to a Gaussian function in the case of triple-axis experiment and to a Lorentzian function in the case of PRISMA data.

data. Such a disagreement is, however, only confined to an energy range where the previous data[12] are scattered around the force-constant curve which has been fitted to them. Of course one cannot rule out the possibility of some instrumental effect, possibly due to the nature of the high-symmetry scan performed in reciprocal space by the PRISMA spectrometer.

In order to ascertain this point some constant-q scans on the same sample have been performed using the more familiar triple-axis spectrometry. The triple-axis spectrometer installed at the Training, Research, and Isotope Production Reactor of Casaccia (Rome) has been employed to collect the data on the transverse-phonon branch along the [00ζ] direction at room temperature. As an example the $\zeta=0.3$ phonon peak at room temperature is shown in Fig. 2 in comparison with similar data recorded on PRISMA, where one observes both the transverse and longitudinal peaks at the same time. We observe that the energy of the transverse branch as measured by triple axis is slightly lower than that determined on PRISMA. Therefore there is an indication that the shift observed in Fig. 1 is at least partly due to a real effect connected to the resolution function of the two spectrometers, although the small number of points used in fitting the dispersion curve also has an effect on the apparent disagreement.

In Fig. 3 we report some of the experimental peaks simply corrected for background and for the incoming spectrum, as a function of temperature. These data show a clear trend in going from 6 to 1.2 K: In particular, the intensity of the longitudinal phonon at about 6 meV is strongly reduced when the temperature is lowered below 4.2 K, whereas no significant change is observed between 6 and 4.2 K. This reduction can be interpreted as due to the broadening of the phonon peak and in any case to an overall shape change of the one-phonon response function. The temperature dependence of the phonon intensity cannot be simply ascribed to a resolution effect. Indeed, in such a case, a complex temperature-dependent energy shift, producing a deformation of the phonon-dispersion curve, should be present. Such a deformation is presently not observed, so that the data are better interpreted in terms of temperature-dependent one-phonon response function.

Finally we mention that the measurements have been repeated several times, both increasing and lowering the temperature between 1.2 and 6 K and for different spectrometer configurations. The use of various spectrometer configurations is helpful in order to identify possible spurious contributions to the scattered intensity, since they should change position in the $(\mathbf{q},\omega)$ space for different spectrometer settings. No such spurious contributions to the scattered intensity have been found in the present energy range. Moreover the observed effect follows closely the temperature changes with no hysteresis for runs lasting from 2 to 12 h.

To obtain a further insight into the low-temperature behavior of Ta, an analysis of the structure of the present sample has been performed using the PRISMA spectrometer as a difractometer. The results have shown that the sample preserves its bcc structure over the whole temperature range and, therefore, no structural phase transition is present in the temperature interval we considered. Further structural data have been obtained using the single-crystal diffractometer SXD, installed at the ISIS spallation neutron source.[15] Two diffraction runs have been performed at 5 and 2.5 K in the $\sin(\theta)/\lambda$ range from 0.6 to 1.6 Å^{-1} on a 1-mm-thick disk obtained from the same cylinder used for the inelastic scans. Integrated intensities and d spacing of several reflections have been measured in order to obtain information on the structural stability as well as on the Debye-Waller factor. The bcc structure is retained in all cases, but the relatively low accuracy of this instrument in determining the lattice parameters prevented us from measuring the lattice parameter with high precision. Nevertheless, within the present error (0.2%) no change of the lattice parameter has been observed between 2.5 and 5 K. The intensity data of 209 reflections measured at 5 K using the SXD spectrometer

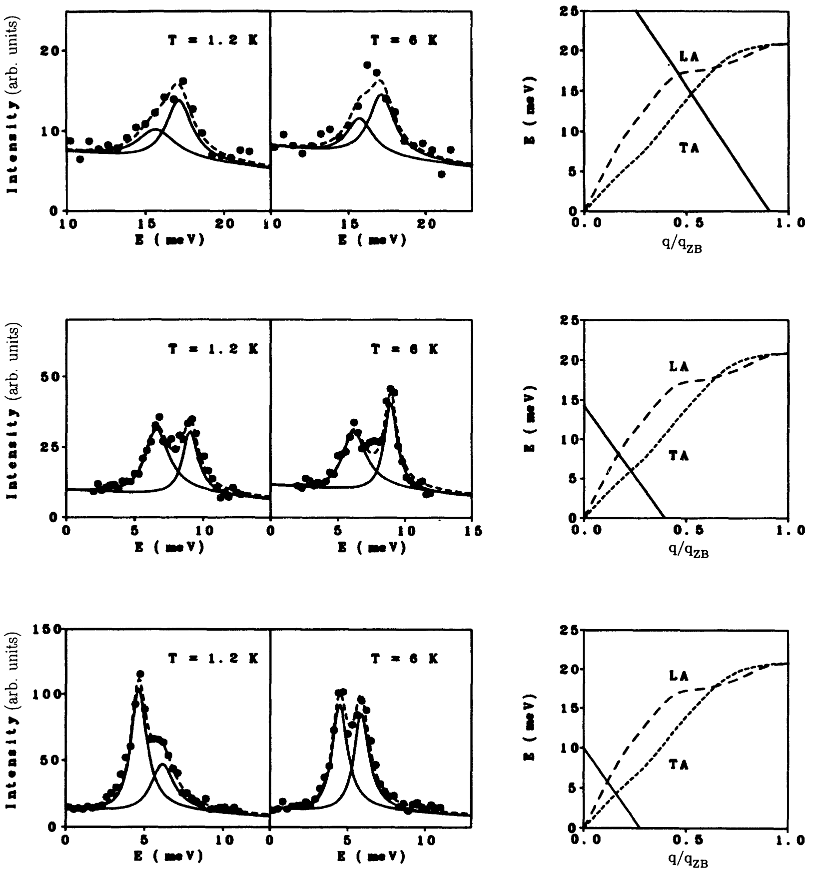

FIG. 3. Selected phonon groups for tantalum at 1.2 and 6 K. Dots: experimental data. Full line: Lorentzian fit of TA and LA phonons to the present data. Dashed line: total fit, as sum of the two components, to the measured intensity. The last two panels on the right side show the dispersion curves for TA and LA phonons as obtained at room temperature from Ref. 12. Also shown is the **q**-E path scanned by the corresponding PRISMA detector.

have been fitted in order to determine the root-mean-square displacement. At 5 K the value $\langle u^2 \rangle = 0.004\,22 \pm 0.00\,76$ Å^2 has been found. This result has to be compared with $\langle u^2 \rangle = 0.004\,86$ Å^2 calculated from the room-temperature density of states.[2] The good agreement between measurement and calculated $\langle u^2 \rangle$ suggests that the low-energy part of the density of states, which is sampled by the present low-temperature experiment, is almost unchanged at low temperature.

III. DISCUSSION

The intensity decrease between 4.2 and 1.2 K observed in the present experiment could be interpreted as due to an increase of the phonon linewidth related to the transition to the superconducting phase which takes place at 4.37 K. Indeed this would have been the case if the superconducting energy gap were about 3 meV as an effect similar to that observed is actually expected when the phonon energy is slightly *larger* than twice the gap.[(6)] However, the isotropic gap in Ta is about 0.7 meV, so that a very large anisotropy is necessary to explain the present data in terms of electron-phonon interactions in the superconducting phase. Alternatively the observed effect could be related to some other kind of transition. We do not think that a magnetic phase transition could be present in Ta, therefore a structural transition could be responsible for the observed behavior. In view of the fact that the diffraction data show that no transition is present at low temperature, the observed phonon damping is more likely due to a structural *instability* rather than to a true transition. A similar situation has been already observed in different systems[16–19] and seems to be related to a structural transition close to the working point in the phase space.

A more general view of the present effect can be gained by analyzing the overall trend of the peak intensities of both longitudinal and transverse-phonon groups versus temperature. In order to get a good estimate of the peak intensity we fitted Lorentzian peaks plus a linear background to the experimental phonon groups. The Lorentzian functions used in the fit were weighted by the temperature factor $[n(\omega)+1]$, $n(\omega)$ being the Bose distribution function. The peak intensity so derived represents a measure of the change of the one-phonon response function, which cannot be determined in the present experiment since the resolution function is not good enough and it is difficult to infer which part of the peak tail is real scattering or background. The whole body of the results obtained by the present fit to longitudinal and transverse-phonon peaks at 1.2, 4.2, and 6 K is summarized in Figs. 4(a) and 4(b). In this figure the ratio of the peak intensity at 1.2 and 4.2 K to that at 6 K is shown versus energy. It is readily seen that a clear decrease of the peak intensity is present for the longitudinal phonons at 1.2 K, while almost no effect is seen at 4.2 K. On the other hand the transverse phonons are scarcely affected at all temperatures.

The effect we observed is rather puzzling because it develops in a temperature range coincident with that of superconductivity. However, the phonon energy involved in this effect is much larger than the superconductivity

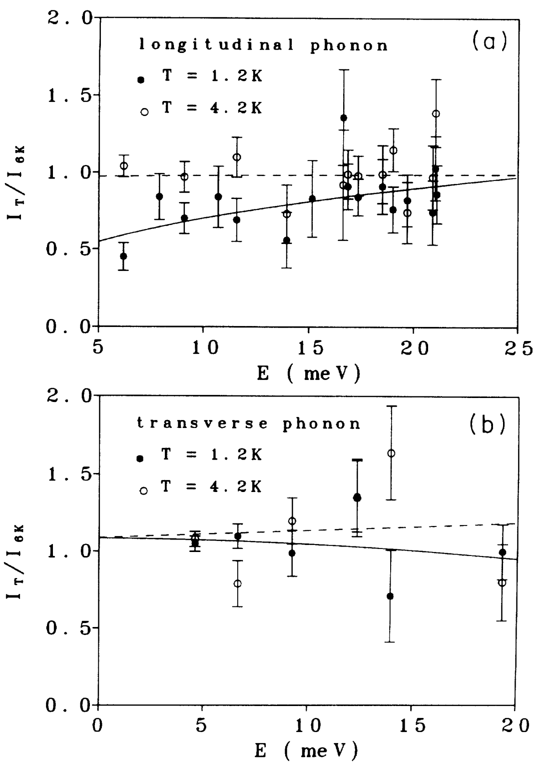

FIG. 4. (a) Ratios of the peak intensity of the longitudinal phonons at temperature $T=1.2$ (dots) and 4.2 K (circles) to that of the corresponding peak at 6 K as a function of the phonon energy. The full and dashed lines are guides to the eye at $T=1.2$ and 4.2 K, respectively. (b) The same as in Fig. 4(a), but for transverse phonons.

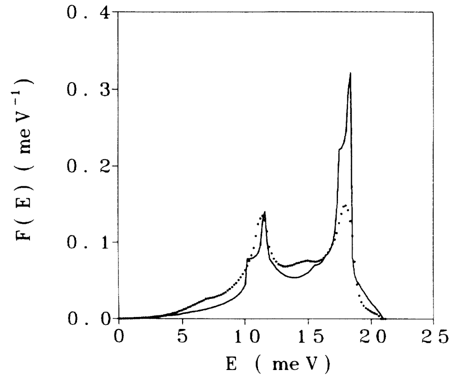

FIG. 5. Density of phonon states in Ta. Full line: unrenormalized density of states from Ref. 12. Dots: tunneling experiment of Ref. 11.

gap so that the correlation between the present effect and the superconducting transition does not appear possible.

In any case interpreting the change of the peak intensity of the longitudinal phonons as related to a broadening of the phonon lineshape, it would be possible to explain the disagreement observed between the phonon density of states measured by the neutron-scattering experiment of Ref. 12 and by the tunneling experiment[11] (see Fig. 5). In fact a broadening of the longitudinal phonon peaks in the low-temperature region would renormalize the phonon density of states given by

$$F(\omega)=\frac{2\omega}{\pi}\Sigma_j\int_{-\infty}^{+\infty}\mathrm{Im}\frac{F_{0j}(\omega')\Gamma_j(\omega')}{[(\omega^2-\omega'^2)^2-\Gamma_j^2(\omega')]}d\omega' , \qquad (1)$$

where the sum runs over the phonon branches. $F_{0j}(\omega)$ is the unrenormalized density of states for the jth branch and $\Gamma_j(\omega)$ is the energy-dependent phonon linewidth. Assuming that from the dispersion relations measured by neutron-scattering experiments the function $F_{0j}(\omega)$ can be derived, Eq. (1) shows that the energy-dependent broadening can produce a reduction of the high-energy peak (around 18 meV, see Fig. 5) of the renormalized density of states with respect to the unrenormalized one. Indeed this peak contains a large contribution from the longitudinal branch. Such an interpretation of the observed behavior could reconcile neutron and tunneling experiments. Nonetheless other experimental investigations are certainly necessary to identify the nature of the presently observed effect.

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