Neutron and microcontact investigations of characteristics of the electron-phonon interaction in technetium

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Inelastic coherent neutron scattering was used to determine the phonon density of states in technetium at temperatures 300 and 150 K. Microcontact spectroscopy was employed to find the electron-phonon interaction function for Tc. The positions of a low-energy peak in the neutron and microcontact spectra differed by 2 meV. This difference was accounted for by assuming that "anomalous" optical phonons played the dominant role in the formation of the microcontact electron-phonon interaction constant.

It is not possible to understand the nature of superconductivity of transition metals without determination of the partial contributions of the separate branches of the phonon spectrum to the electron-phonon interaction (EPI) function. These contributions can be found only if detailed information on the phonon dispersion law and on the Fermi surface of the investigated metal is available. Since such full information is still inaccesible, it is usual to describe the superconducting properties of metals in terms of the Éliashberg EPI function $G(\omega) = \alpha^2(\omega)F(\omega)$ [$F(\omega)$ is the density of the phonon states and $\alpha^2(\omega)$ is the square of the matrix element of the EPI averaged over the Fermi surface]. The phonon states density $F(\omega)$ can be found experimentally from neutron scattering experiments, whereas $G(\omega)$ can be deduced by tunnel measurements or, accurate to within the form factor, from microcontact spectroscopy. Use of this function makes it possible to describe the situation which applies to nontransition metals. In the case of transition metals a wide range of experimental results obtained in studies of the EPI can also be described in the framework of Éliashberg's theory but there are cases when this approach is clearly unsuitable. For example, experimental studies of the electron damping in Nb_3Sn (Ref. 2) and Nb (Ref. 3) as well as invesigations of anomalies of the phonon dispersion curves of some transition metals, and of their alloys and compounds,[4,5] indicate that in a certain part of the phase space the EPI may become much stronger for some branches of the phonon spectrum.[6,7] As a rule, the presence of anomalies of this type is correlated with a high critical temperature. However, experimental determinations of the dispersion curves provide information only on the behavior of one or several branches of the phonon spectrum and do not answer the question of the contribution of "anomalous" branches to the total density of phonon states and to the EPI function.

The special properties of technetium suggest that this metal may exhibit anomalies of the dispersion curves in the behavior of the integral functions $F(\omega)$ and $G(\omega)$. Technetium differs from other transition metals with the hcp structure particularly because of its high temperature T_k (8.2 K) and a strong anomalous "dip" exhibited near the zone center by the dispersion curve of longitudinal optical phonons propagating along the hexagonal axis (LO [001]).[8] Smith and Vakabayashi[9] demonstrated that this branch exhibits also an anomalous temperature dependence: cooling from $T = 300$ K to $T = 50$ K reduces the frequency near the zone center from 16.5 to 10.5 meV. These characteristics can be explained[9] by a strong interaction of the LO [001] phonons with the electron subsystem.

The purpose of the present investigation was to estimate experimentally the contribution of anomalous phonon branches of technetium to $F(\omega)$, firstly by comparing the results of neutron investigations carried out at various temperatures and secondly by determining their contribution to the EPI function from a comparison of neutron and microcontact measurements.

Microcontact measurements were carried out both on bulk polycrystalline samples ($R_{300}/R_{10} = 20$) and on films ($R_{300}/R_{10} = 3.5$) prepared by the evaporation of technetium[10] of $d = 1000$ Å thickness on metal (silver) substrates. This Tc film thickness excluded a possible influence of the metal electrode (Ag) on the microcontact spectra. Measurements were made at $T = 4.2$ K in a field of $H = 45$ kOe, which made it possible to study technetium in the normal state. The usual modulation method was employed to determine the second derivative of the current-voltage characteristic d^2I/dV^2 of a point contact known to be proportional to the microcontact function of the EPI $G_{pc}(\omega)$. A point contact between two electrodes was produced by the standard shear method.

Figure 1 shows typical microcontact spectra of bulk
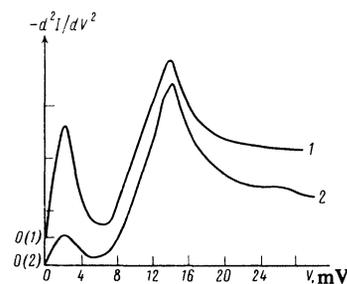
FIG. 1. Second derivative of the current-voltage characteristic obtained for bulk (curve 1) and film (curve 2) samples. Parameters: 1) $R = 5$ Ω, $U_{mod} = 1$ mV, $H = 45$ kOe, $T = 4.2$ K; 2) $R = 1$ Ω, $U_{mod} = 1$ mV, $H = 45$ kOe, $T = 4.2$ K. The maximum of the second derivative is located at $V = 14.0 \pm 0.5$ mV.
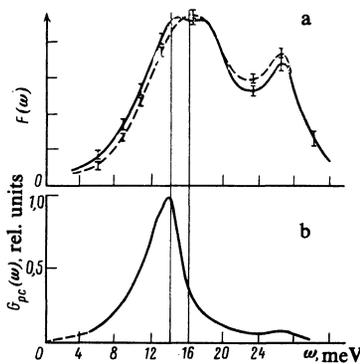
FIG. 2. a) Density of phonon states in technetium at $T = 300$ K (dashed curve) and at $T = 150$ K (continuous curve). b) Microcontact EPI function $G_{pc}(\omega)$ of a film sample after subtraction of the background.

(curve 1) and film (curve 2) samples. The quality of the spectra obtained for film samples was better than that of bulk crystals. This was manifested particularly by the higher intensity of the spectra, the lower level of zero-point anomalies (characteristics of the microcontact spectra at energies up to 5 meV), and the lower background level. This was probably due to the fact that the cleanliness of the film surfaces formed in ultrahigh vacuum was higher than that of bulk samples. Clearly, this was also the reason for the absence of a high-energy peak in the spectra of the bulk samples.

Neutron measurements were made employing a time-of-flight spectrometer with a cold neutron source.[11] A sample was in the form of a Tc foil $d = 0.2$ mm thick. Figure 2a (dashed curves) gives the results of neutron investigations at room temperature. The phonon spectrum had two clear maxima: a wide one centered on the energy $\omega = 16.0 \pm 0.5$ meV and a narrow one at $\omega = 27.0 \pm 1.0$ meV. The peak positions were in good agreement with the data deduced from dispersion curves,[8] but the limiting value of the frequency obtained in the present study was somewhat higher (30 and 25 meV, respectively).

We shall compare the results of neutron (at $T = 300$ K) and miscrocontact measurements (Fig. 2). Figure 2b shows the microcontact spectrum of a film sample with the background subtracted. This subtraction was carried out by a method described in Ref. 12. Firstly, we can see that the microcontact spectra are characterized by a much lower ratio of the intensities of the high-energy (G_2) and low-energy (G_1) peaks compared with the corresponding ratio in the case of the neutron spectra. Usually the reduction in the relative intensity of the peak G_2 in the microcontact spectra is attributed to the heating of the contacts by the current and the associated broadening of the spectral lines,[13] as well as to a considerable plastic deformation of the metal in the contact region.[14] In the case of transition metals this reduction may also be a consequence of the energy dependence of ε^2 (Ref. 1). Secondly, there is a clear displacement of the peak in the microcontact spectrum toward lower energies by an amount 2–2.5 meV. Such a considerable difference between the positions of the $\alpha^2(\omega)F(\omega)$ and $F(\omega)$ peaks has not been observed earlier for any one of the large number of the investigated materials. The reason for this difference may be the change

in the phonon spectrum of Tc with temperature due to (as already pointed out) the anomalous temperature dependences of the individual phonon branches. Moreover, it is reported in Refs. 15 and 16 that the temperature dependences of the ratio of the lattice constants c/a and of the Debye temperature Θ_D of rhenium, which is a homolog of technetium in the periodic system, are unusual. For this reason it is correct to compare the microcontact spectra with the neutron spectra at low temperatures. It follows from the results of Ref. 9 that the main softening of the LO [001] mode occurs in the temperature interval from $T = 300$ K to $T = 150$ K (the frequency near the center of the zone then changes from $\omega = 16.5$ meV to $\omega = 12$ meV).

The density of states $F(\omega)$ determined at $T = 150$ K is given in Fig. 2a (continuous curve); both the continuous and dashed curves are normalized to the same area.

Softening of the low-energy spectrum as a result of cooling is evidence of a considerable contribution of the anomalous optical phonons to $F(\omega)$, but this contribution does not shift significantly the low-energy peak in the distribution $F(\omega)$. In comparing the microcontact and neutron spectra at $T = 150$ K (Fig. 2) we can see that the difference between the peak positions is still considerable (2 meV). In our opinion, this difference is an indication of the dominant role of the anomalous phonons in the formation of the microcontact EPI function.

The phonons near the zone center (characterized by a small transferred momentum) can make a considerable contribution to the EPI function only via umklapp processes. The possibility of occurrence of these processes in technetium is suggested by the results of an investigation of the de Haas-van Alphen effect[17] and by theoretical calculations of the energy band structure.[18]

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