INVESTIGATION OF THE PHONON SPECTRUM OF VANADIUM

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The spectrum of neutrons scattered inelastically from a vanadium sample was measured using a time-of-flight neutron spectrometer. A digital computer was used to reconstruct the phonon spectrum from the experimental data and the accuracy of this reconstruction was determined.

1. INTRODUCTION

THE phonon spectrum $g(\omega)$ is an important quantity for solid state physics. Until recently the main method for finding the form of the phonon spectrum was the calorimetric method in which one studies the temperature dependence of the specific heat at constant volume. However, it has been shown theoretically[1,2] that the reconstruction of the spectrum of excitations of a Bose system from its specific heat, which involves the solution of an integral equation with an unstable kernel, cannot be done uniquely with the present-day calorimetric technique.

The possibility of a direct determination of the spectrum $g(\omega)$ from experimental data arose only recently in connection with the start of studies of inelastic scattering of "cold" and slow neutrons and the Mössbauer effect. In these experiments the measured quantities are linear functions of $g(\omega)$, and the problem of reconstructing the phonon spectrum is simplified considerably.[3-6]

One of the first substances for which inelastic scattering of neutrons was studied in order to determine $g(\omega)$ was vanadium, which is a monoisotopic metal with a body-centered cubic lattice.[7-10] Because of spin incoherence, its coherent scattering cross section for neutrons is negligibly small, so that one measures the cross section for inelastic incoherent scattering which is simply related to the function $g(\omega)$.[3] The main features of the data on the vanadium spectrum, as determined from neutron measurements, are shown in [8,9], but the relation between the ordinates of the maxima of the phonon spectrum and their widths are somewhat contradictory. We therefore made a new measurement of the spectrum from inelastic scattering of "cold" neutrons by vanadium, using a neutron time-of-flight spectrometer with a resolution higher than that used previously, and analyzed the accuracy of the corrections made in reconstructing the phonon spectrum.

2. MEASUREMENT PROCEDURE

The measurements of the spectrum of inelastically scattered neutrons were made on equipment set up at the IRT-1000 reactor.[11] The vanadium sample, in the form of a plate $15\times18\times1$ cm was placed in the scattering chamber at an angle of 45° to the incident beam. The sample temperature was 293°K and the vanadium content in the sample exceeded 99.7%. The spectrum of scattered neutrons was measured over the energy range from 10^{-2} to 5×10^{-2} eV at an angle of 90° to the incident beam. The chopper had a slit width $s = 8\times10^{-2}$ cm, the speed of revolution of the chopper rotor was 4500 rpm and the width of the channels of the time analyzer was 32 μsec. With our choice of rotation speed of the chopper, the maximum of the transmission function corresponded to approximately the middle of the energy range studied, and dropped to 0.5 at the ends of the range. Spectral measurements at energies below 10^{-2} eV were of no interest since this part of the scattering curve can easily be computed from the elastic constants of vanadium[12] and contains no additional physical information.

The measurements of the spectrum of inelastically scattered neutrons were made with the reactor operating at nominal power, and the intensity of scattered neutrons was 0.6 neuts/min per channel with a background of 0.3 neuts/min. Thus more than 100 hours of measurements were needed to get satisfactory statistical accuracy.

Since the chopper of the time-of-flight spectrometer has parabolic slits and operates as a crude monochromator, to obtain the true spectrum of inelastically scattered neutrons one must correct the primary experimental data for the deformation of the spectrum by the transmission function of the chopper. For the chopper used in this experiment, the transmission function was deter-

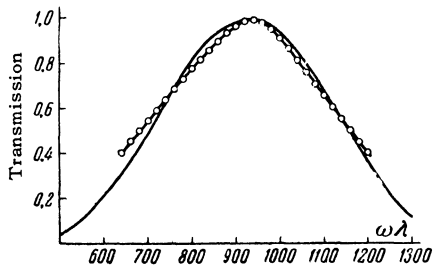

FIG. 1. Comparison of experimentally determined and computed chopper transmission functions.

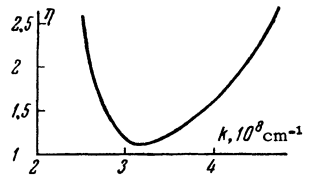

FIG. 2. Dependence of total correction η on wave number of scattered neutron.

mined experimentally. Figure 1 shows the results and a comparison with the theoretical transmission function.

In addition one must take account of deviations of detector efficiency from a 1/v law and of attenuation of the neutrons in the air along the path between the chopper and the detector, where the path length is increased because of the scattering. Figure 2 shows the dependence of the total correction η on the wave number of the scattered neutron, including all three factors, while Fig. 3 shows the experimental curve of the spectrum of inelastically scattered neutrons when all the corrections are included. As we see from Fig. 3, the maxima in the spectrum of scattered neutrons are well resolved and the maximum at higher energy is the more intense one.

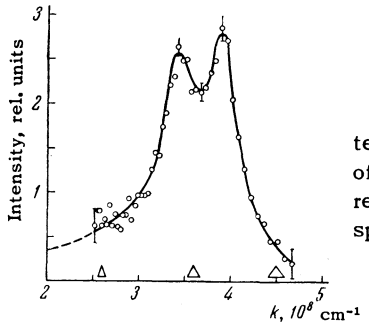

FIG. 3. Intensity of scattered neutrons as a function of wave number; Δ is the resolution of the neutron spectrometer.

3. RECONSTRUCTION OF THE PHONON SPECTRUM

As already mentioned, for vanadium the probability of scattering of a neutron with absorption or emission of one phonon of frequency ω is proportional to the density of the phonon spectrum at the given frequency:[3]

$$W^{(\pm 1)} = F(E, \mathbf{n}, E', \mathbf{n}')\, g(E - E'), \qquad (1)$$

where $F(E, \mathbf{n}, E', \mathbf{n}')$ is a factor which contains functions specific for the scattering as well as simple normalization factors: E and $\mathbf{n}$ are the energy and unit vector along the direction of the scattered neutron; E′ and $\mathbf{n}'$ are the corresponding quantities for the incident neutron. Roughly speaking, the experimental spectrum of inelastically scattered neutrons (Fig. 3) enables one, using (1), to determine directly the phonon spectrum of the material.

However, an analysis made in this approximate way does not permit the reconstruction of the function $g(\omega)$ with the maximum attainable accuracy for this experiment. Actually the expression (1) is strictly applicable only to the spectrum of neutrons scattered by an infinitely thin sample, where the contribution from multiphonon scattering has been subtracted, and applies only to an apparatus with infinite resolution. These requirements are not met in the experiment. Thus to reconstruct $g(\omega)$ from the neutron experiment one must consider the effect of distortion of the spectra of incident and scattered neutrons due to target thickness, the contribution of multiphonon and successive one-phonon processes, and also include the effect of non-monochromaticity of the primary spectral line of "cold" neutrons and the finite resolution of the time-of-flight spectrometer, which determine the overall resolution of the equipment.

It is convenient to carry out the procedure for reconstructing $g(\omega)$ in two stages: 1) reduction of the experimentally measured spectrum of inelastically scattered neutrons to the spectrum for an infinitely thin sample and subtraction of the contributions from multiphonon and successive one-phonon processes; 2) reconstruction of the function $g(\omega)$ from the spectrum for the infinitely thin sample taking into account the overall resolution of the apparatus.

The complete solution of the problem of finding $g(\omega)$ for vanadium was obtained using the TsM-2 digital computer. Figure 4 shows the function $\tilde{g}(\omega)$ obtained in the first stage of the reconstruction, while Fig. 5 gives the final result. In the interval from $\omega = 0$ to $\omega_1 = 1.52 \times 10^{13}$ sec^{-1}, the function $g(\omega)$ is given by $B\omega^2$ (with $B = 0.0579$), while in the range $\omega > \omega_1$ it is reproduced by the expansion

$$g(\omega) = c \sum_{\alpha=0}^{13} g_\alpha f_\alpha(\omega) \qquad (2)$$

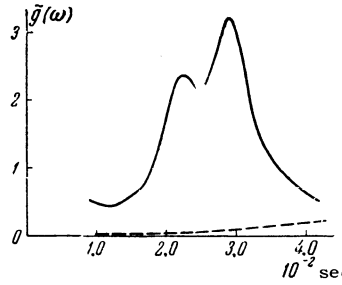

FIG. 4. Phonon spectrum of vanadium, constructed assuming a delta-function shape for the overall resolution function. The dashed curve gives the contribution of twofold and two-phonon processes.

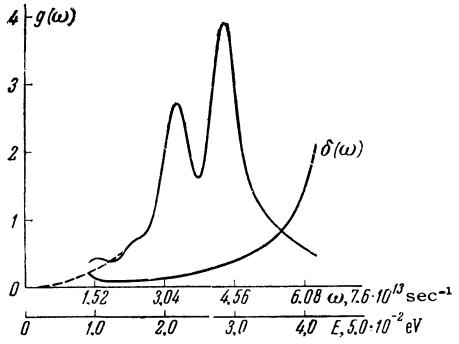

FIG. 5. Phonon spectrum of vanadium, constructed using the overall resolution function. The curve $\delta(\omega)$ indicates the accuracy in reconstructing the spectrum. The dashed curve shows the low-frequency part of the Debye spectrum for $\Theta = 338°$ K.

in the system of basis functions $f_\alpha(\omega)$, defined by the relation

$$f_\alpha(\omega) = \sum_{\beta=0}^{\alpha} f_{\alpha\beta} \cos[\pi\beta(\omega - \omega_0)/\Omega], \qquad (3)$$

where α and β are integral-valued, and g_α and $f_{\alpha\beta}$ are expansion coefficients, ω_0 is the lower limit of the spectral region investigated, which was 0.714×10^{13} sec^{-1}, Ω gives the entire frequency range over which the reconstruction was done, and c is a normalization coefficient, equal to 0.0218.

The expansion coefficients found in the reconstruction are shown in the table. Representing the phonon spectrum of vanadium by this kind of expansion is very convenient since it enables one to compute quantities (like specific heat, Debye-Waller factor, etc) which depend on the phonon spectrum, and also to construct the dispersion curves from the data on the phonon spectrum of the vanadium lattice. In Fig. 5 we show, together with $g(\omega)$, the error curve $\delta(\omega)$ for the reconstructed function:

Expansion coefficients of $g(\omega)$

α	0	1	2	3	4	5	6	7	8	9	10	11	12	13
$g(\alpha)$	62.04	1.34	−26.25	−2.10	5.91	1.89	4.24	1.98	−5.21	−2.20	3.77	1.75	−1.47	−0.55

$f_{\alpha\beta}$

β / α	0	1	2	3	4	5	6	7	8	9	10	11	12	13
0	0.0432													
1	−0.0222	0.0786												
2	0.0241	−0.0217	−0.0855											
3	0.0004	0.0529	−0.0234	0.0855										
4	0.0284	−0.0007	0.5530	−0.0280	0.0917									
5	−0.0001	0.0612	−0.0021	0.0627	−0.0305	0.1025								
6	0.0338	−0.0006	0.0699	−0.0027	0.0756	−0.0357	0.1131							
7	−0.0005	0.0786	−0.0010	0.0844	−0.0055	0.0886	−0.0460	0.1304						
8	0.0465	−0.0009	0.0962	−0.0034	0.0996	−0.0129	0.1101	−0.0588	0.1506					
9	−0.0006	0.1143	−0.0039	0.1151	−0.0109	0.1235	−0.0230	0.1364	−0.0793	0.1792				
10	0.0712	−0.0013	0.1400	−0.0084	0.1454	−0.0180	0.1545	−0.0368	0.1747	−0.1036	0.2195			
11	−0.0018	0.1751	−0.0076	0.1772	−0.0176	0.1833	−0.0337	0.1976	−0.0604	0.2312	−0.1460	0.2704		
12	0.1085	−0.0105	0.2218	−0.0174	0.2245	−0.0340	0.2358	−0.0575	0.2607	−0.0999	0.3068	−0.2080	0.3457	
13	−0.0108	0.2780	−0.0251	0.2836	−0.0384	0.2917	−0.0629	0.3126	−0.1023	0.3494	−0.1671	0.4270	−0.3083	0.4540

$$\delta(\omega) = \left\{\sum_{\alpha=0}^{13} f_\alpha^2(\omega)\right\}^{1/2}, \quad (4)$$

and the dashed curve gives the low-frequency part of the function $g(\omega)$ computed assuming a Debye characteristic temperature $\Theta = 338°K$ for vanadium. The limitation of the expansion to 14 harmonics is related to the requirement of "smoothness" of the error curve $\delta(\omega)$, which was imposed on the solution of the fitting problem.

DISCUSSION OF RESULTS

A comparison of the results for $g(\omega)$ with those of Eisenhauer et al [9] and Stewart and Brockhouse [8] shows that the phonon spectrum given in Fig. 5 is closer to that of Eisenhauer et al. The energy positions of special points of the spectrum are the same. But there is a considerable sharpening up of the maxima and a change in the relation between their ordinates. The maximum corresponding to higher frequencies has increased in height. Such a change in the character of the phonon spectrum is evidence for a much greater influence of noncentral interactions than one would have expected on the basis of the work of Stewart and Brockhouse [8] and Eisenhauer, [9] since the phonon spectrum differs even more markedly from that predicted from computations for body-centered lattices with central interaction. [13,14] This result should be regarded as a natural one for vanadium, since it is a transition metal in which an important part is played by covalent bonds, which are highly noncentral. A more detailed discussion of the characteristics of the interatomic forces can be given only after the phonon dispersion curves are constructed.

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