

Testing the phonon spectrum of metallic chromium with the nuclear-resonance photon scattering technique

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The nuclear-resonance photon-scattering cross section σ_s from ^{50}Cr in a metallic form was studied as a function of temperature in the range 15–300 K. The value of σ_s was used for deducing the effective temperature T_e which is related to a weighted average of the phonon spectrum of metallic Cr. This T_e is also a measure of the total zero-point kinetic energy of the lattice and was used to define another Debye temperature Θ_0 ; its value for metallic Cr was measured and found to be $\Theta_0=510\pm 20$ K. This value is higher by about 10% than that obtained from the known phonon spectrum deduced by inelastic neutron scattering. A similar deviation occurs in other elements. The reason for this deviation of the values of T_e and hence of Θ_0 obtained by the nuclear-resonance photon-scattering technique and by inelastic neutron scattering is discussed.

The nuclear-resonance photon-scattering (NRPS) technique¹⁻⁴ was used for studying the effective temperature T_e and the Debye temperature Θ_0 of metallic Cr. The concept of an effective temperature was introduced by Lamb⁵ to take into account the vibrational motion of the atoms in the lattice; it is expressed by

$$T_e = \frac{1}{k} \int_0^\infty h\nu g(\nu) \left[\frac{1}{\exp(h\nu/kT) - 1} + \frac{1}{2} \right] d\nu, \quad (1)$$

where $g(\nu)$ is the real phonon spectrum measured by inelastic neutron scattering (INS). In case where the lattice is described by a Debye-type phonon spectrum

$$g(\nu) = 3\nu^2/\nu_m^3 \quad (2)$$

with ν_m the maximum cutoff frequency, related to the Debye temperature of the solid, $\Theta_0 = h\nu_m/k$, Eq. (1) takes the form

$$T_e/T = 3(T/\Theta_0)^3 \int_0^{\Theta_0/T} x^3 \left[\frac{1}{\exp(x) - 1} + \frac{1}{2} \right] dx \quad (3)$$

with $x = h\nu/kT$. The limiting value of T_e at $T=0$ K is

$$T_0 = (3/8)\Theta_0. \quad (4)$$

T_0 can be deduced from Eq. (1) because the real phonon spectrum $g(\nu)$ is usually known.⁶ Equation (4) may be viewed as another definition of the Debye temperature which we call the INS Debye temperature. Equation (3) can also be used to calculate Θ_0 as a function of T where T_e is taken from Eq. (1). Figure 1 is a plot of $\Theta_0(T)$ deduced in this manner where $g(\nu)$ for metallic Cr is taken from Ref. 6; it may be seen to be practically independent

of T . This Θ_0 is very useful not only in NRPS work but also in neutron resonance research because it yields a precise value of the Doppler width of the nuclear level (see below). It should also be added that the concept of a Debye temperature Θ_D does not mean that we have resorted to the Debye approximation; it is just a convenient way to express $\langle \nu g(\nu) \rangle$ through Eqs. (1) and (3). In the case of a strict Debye model for $g(\nu)$, the value of Θ_0 reduces to the commonly used "caloric" Debye temperature Θ_D derived from the lattice specific heat C_p . Here we remind the reader that C_p in the harmonic approximation may

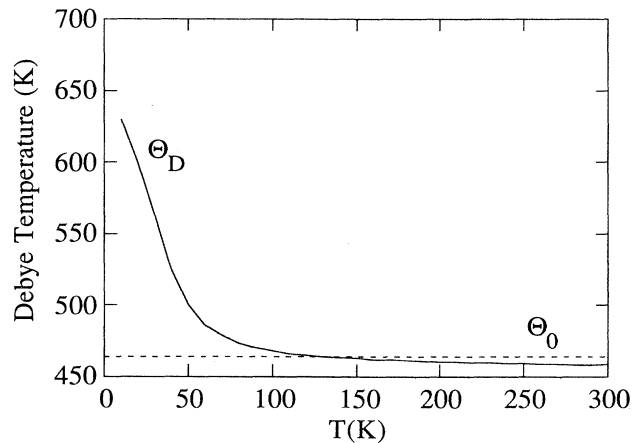


FIG. 1. Debye temperatures Θ_0 and Θ_D as a function of T calculated using the real phonon spectrum $g(\nu)$ of metallic Cr taken from Ref. 6.

be deduced from the entropy of the lattice yielding

$$C_p = R \int_0^\infty g(\nu) [(e^x - 1)^{-1} + 1] (e^x - 1)^{-1} x^2 d\nu \quad (5)$$

with $x = h\nu/kT$. By inserting the real phonon spectrum $g(\nu)$, the specific heat C_p may be calculated as a function of T . For a Debye solid $g(\nu)$ may be replaced by Eq. (2) yielding

$$C_p' = 3R (T/\Theta_D)^3 \int_0^{\Theta_D/T} x^4 e^x (e^x - 1)^{-2} dx. \quad (6)$$

Thus Θ_D is defined by requiring

$$C_p(T) = C_p'(T). \quad (7)$$

A plot of $\Theta_D(T)$ is shown in Fig. 1 and may be seen to be strongly dependent on T and differs markedly from the behavior of $\Theta_0(T)$. The present value of Θ_0 gives a measure of the total zero-point kinetic energy of the atoms in the lattice, and hence of $\langle v^2 \rangle$, it is sensitive chiefly to the optical high-frequency phonon modes as is evident from Eq. (1); it differs from the x-ray measurements which gives a measure of $\langle z^2 \rangle$ and is more sensitive to the acoustical low-frequency modes. In fact, in lattices where $\langle v^2 \rangle$ is small the value of $\langle z^2 \rangle$ is expected to be large and a good accuracy in Θ_0 is obtained with x-ray measurements, while in cases where $\langle z^2 \rangle$ is small, the value of $\langle v^2 \rangle$ is expected to be large and a good accuracy in Θ_0 is obtained with the NRPS method. In a sense the two techniques complement each other. Besides the above definitions of Debye temperature, various other definitions are used in the literature.⁷⁻⁹ In the present work we will refer mainly to the INS and the "caloric" Debye temperature defined above.

It was shown that under certain conditions, the NRPS technique can be used for the determination of T_e of some elements either in a metallic form^{1,2} or in other chemical forms.^{3,4} Such a measurement is very useful because it can provide a weighted average of the phonon spectrum; this can be compared with that calculated from INS data using numerical integration thus testing the validity of such spectra. A scan of previous measurements¹⁻⁴ has shown that T_e of metallic elements, deduced using the NRPS technique, is higher by about 10% than the value which could be deduced from INS data. The purpose of the present work is twofold. First, to find a precise value of T_e and Θ_0 because these are useful parameters in analyzing NRPS data where metallic Cr is used as a γ source.⁴ In this connection it may be added that, in the literature, the reported Θ_D values of Cr differ markedly from each other and are in the range 400 to 630 K. Second, to test the validity of the known phonon spectrum of metallic Cr obtained by INS and to try to explain the 10% deviation which seem to recur in the present case.

The possibility of measuring T_e by NRPS depends on the occurrence of a random overlap¹⁰ between the 8888-keV γ -ray line emitted by the $^{54}\text{Fe}(n, \gamma)$ reaction and a nuclear level in ^{50}Cr . This random photoexcitation process in Cr was first reported by Moreh, Shlomo, and Wolf¹⁰ where the spectroscopic properties of the resonance level were determined: the spin $j=1$, the ground-state spin $J=0$, the total radiative width $\Gamma=0.75 \pm 0.20$

eV, the ground-state branching ratio $\Gamma_0/\Gamma=0.90 \pm 0.07$, and the energy distance between the incident- γ -ray line and the resonance level, $\delta=18 \pm 1$ eV. The Doppler-broadened shapes of the incident- γ -ray line and the resonance level are illustrated in Fig. 2. It turns out that the sensitivity of the present method for the determination of T_e and hence of Θ_0 depends primarily on the relative magnitudes of δ and the Doppler widths Δ_s and Δ_r of the incident γ -ray line and the resonance level. The accidental large value of δ enabled us to determine T_e by making a detailed study of the scattering cross section as a function of T .

From Fig. 3 [which shows a plot of the ratio of the scattering cross sections $R_T = \sigma_s(10 \text{ K})/\sigma_s(297 \text{ K})$ against δ] it may be seen that if $\delta \approx 0$, then the value of σ_s differs from 1 only slightly. In addition a large variation of σ_s with T , and hence good accuracy in the determination of T_e is obtained for cases where (i) $\delta > \Delta_s, \Delta_r$ and (ii) $\Delta_r > \Gamma$. Moreover, for $\delta > \Delta_s, \Delta_r$, the value of R_T can differ markedly from unity and hence a measurement of R_T can determine T_e with good accuracy. Furthermore, if Δ_r is small relative to Γ , then the shape of the resonance level will be determined primarily by the value of Γ and only slightly by Δ_r or T . The above points may further be illustrated by considering Fig. 4 which shows a plot of the uncertainty in the deduced value of Θ_0 resulting from a 1% error in the experimental measurement of the value of R_T . Here again the uncertainty may be seen to decrease with increasing value of δ .

Theoretically, the Doppler widths Δ_s, Δ_r are defined by^{11,12}

$$\Delta_i = E_i (2kT_i / M_i c^2)^{1/2} \quad (8)$$

with ($i=r,s$); E_i , T_i , and M_i are the corresponding energies, effective temperatures, and masses of the resonance

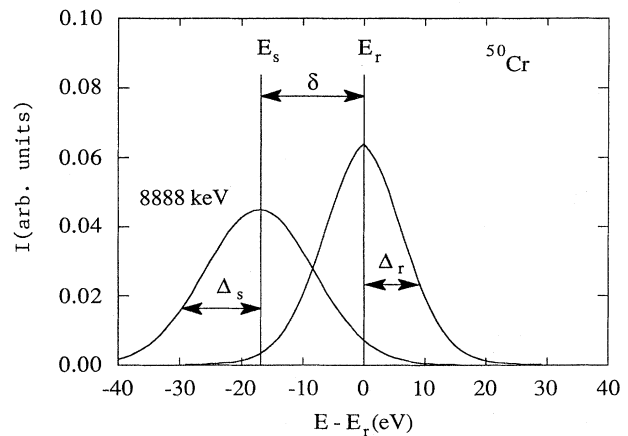


FIG. 2. Calculated shapes of the Doppler-broadened incident γ -ray line originating from the $^{54}\text{Fe}(n, \gamma)$ reaction of peak energy E_s (described by a Gaussian having $\Delta_s=11.97$ eV with a γ -ray source temperature of 498 K) and a Doppler-broadened 8888-keV nuclear level in ^{50}Cr of peak energy E_r (described by a ψ function with $\Delta_r=9.99$ eV at $T=297$ K) separated by $\delta=17$ eV. The overlapping area is related to the scattering cross section σ_s .

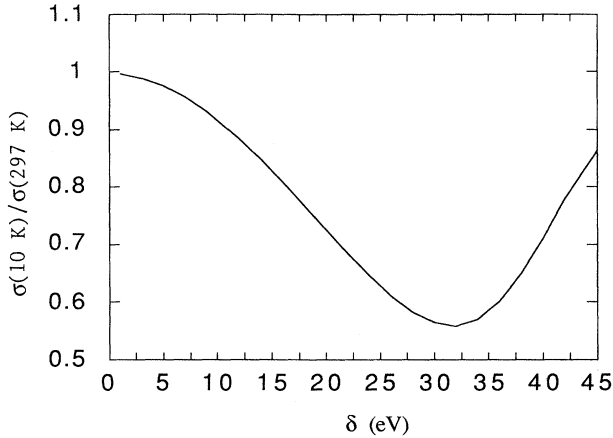


FIG. 3. Calculated ratios of scattering cross sections at $T=10\text{ K}$ and $T=297\text{ K}$ as function of δ from the 8888-keV level of ^{50}Cr . The parameters used were $\Gamma=0.75\text{ eV}$, $\Gamma_0/\Gamma=0.9$, and $\Delta_s=11.97\text{ eV}$. The assumed values of Θ_0 of metallic ^{50}Cr and the iron γ -ray source were 520 and 460 K, respectively. The method of calculating the scattering cross section was given in Ref. 1.

scatterer and the emitting γ -ray source. The shape of the Doppler-broadened γ -ray line emitted by the $^{54}\text{Fe}(n,\gamma)$ reaction, is represented by a Gaussian^{4,11}

$$F(E)=(1/\Delta_s\gamma\pi)\exp[-(E-E_r+\delta)^2/\Delta_s^2]. \quad (9)$$

The shape of the Doppler-broadened nuclear resonant level in ^{50}Cr is a ψ function which is a convolution between a Breit-Wigner resonance form and a Gaussian distribution of energies^{4,11}

$$\sigma_s(E)=\sigma_0\psi(x,t), \quad (10)$$

where $\sigma_0=\lambda^2g\Gamma_0/2\pi\Gamma$ is the maximum cross section for a nonbroadened nuclear level whose ground-state width is Γ_0 and $g=(2j+1)/(2J+1)$ is a statistical factor. The ψ function is given by^{11,12}

$$\psi(x,t)=[1/2(\pi t)^{1/2}]\int_{-\infty}^{\infty}\frac{\exp[-(x-z)^2/4t]}{1+z^2}dz \quad (11)$$

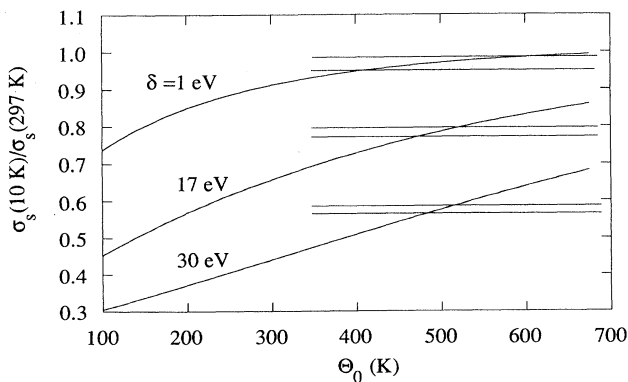


FIG. 4. Calculated values of R_T vs Θ_0 (the Debye temperature of metallic Cr) for three different values of δ . Hypothetical 1% errors in the experimental measurement of R_T are indicated and are found to lead to vastly different uncertainties in the deduced values of Θ_0 ; these are ± 95 , ± 23 , and $\pm 12\text{ K}$ corresponding to $\delta=1$, 17, and 30 eV, respectively.

with $x=2(E-E_r)/\Gamma$, $t=(\Delta_r/\Gamma)^2$, and $\Delta_r=E_r(2kT_r/M_r c^2)^{1/2}$. Both Doppler-broadened shapes are depicted to scale in Fig. 1. The scattering cross section is given by the overlap integral of the above two line shapes; it may be evaluated to yield¹¹

$$\sigma_s=\sigma_0\int_0^\infty F(E)\psi(x,t)dE=\sigma_0\psi(x_0,t_0), \quad (12)$$

where $x_0=2|E_r-E_s|/\Gamma=2\delta/\Gamma$, and $t_0=(\Delta_s^2+\Delta_r^2)/\Gamma^2$. The scattered intensity may be calculated by taking into account the target thickness, the specific geometry of the target-detector-beam system and employing Eq. (12). More details concerning the calculation of the scattered intensity is given in Ref. 1.

The photon source was obtained from the (n,γ) reaction on four metallic iron discs. For measuring the scattered intensity versus T , a 12.7 cm \times 12.7 cm NaI cylindrical detector was used, placed at a distance of 25 cm from the target, and making an angle of 130° with the photon beam. A 160-g rectangular metallic sample of chromium (7.5 cm \times 4 cm) was placed perpendicular to the beam and positioned inside a Displex cryostat which varied the temperature of the target between 12 to 297 K. The running time for each data point was about 12 h. Details of the experimental system are given in Ref. 4.

The experimental scattered intensities I versus T for Cr are shown in Fig. 5 and are normalized so that $I(297\text{ K})=1$. At each point the background contribution was subtracted by using an iron scatterer.

The determination of Θ_0 was made by searching for the values of both δ and Θ_0 which best fitted the data points of Fig. 5. To do so, the elastically scattered intensity was calculated as a function of T using the parameters of the resonance level mentioned above and listed in the captions of Figs. 2 and 3. It should be noted that the Doppler width, $\Delta_s=11.97\text{ eV}$, of the 8888-keV γ -ray line emitted by the $^{54}\text{Fe}(n,\gamma)$ reaction corresponds to an effective temperature $T_s=539\text{ K}$. This value of T_s was obtained at an iron source temperature $T=518\text{ K}$ and using $\Theta_0(\text{Fe})=460\text{ K}$ for metallic iron. We thus obtained $\Theta_0(\text{Cr})=520\pm 20\text{ K}$, $\delta=17.0\pm 0.5\text{ eV}$. The δ obtained here is more precise than that reported previously¹⁰ ($\delta=18\pm 1.0\text{ eV}$) which was based on measuring the cross-section ratio between $T=78$ and 297 K only. This Θ_0 corresponds to an effective temperature (at 0 K) of $T_0=195\text{ K}$ and is characteristic of the ^{50}Cr isotope. The latter value should be compared with $T_0=177\text{ K}$ (and $\Theta_0=473\text{ K}$) deduced from Eq. (1) using the phonon spectrum $g(\nu)$ of metallic natural chromium,⁶ after introducing an $M^{-1/2}$ correction for the isotopic effect as explained below. The calculated scattering intensity ratios obtained using $\Theta_0=473\text{ K}$ is given in Fig. 5 which illustrates the extent of the deviation obtained when using the INS value of Θ_0 .

The isotopic mass correction may be effected by noting that the above values of Θ_0 are related to ^{50}Cr (whose atomic mass is 49.946 amu) while the literature values correspond to a natural isotopic composition whose average mass is 51.996. Applying an $M^{-1/2}$ correction, we obtain for the NRPS value of metallic natural Cr $\Theta_0=510\pm 20\text{ K}$. Higher values of Θ_0 as compared to the

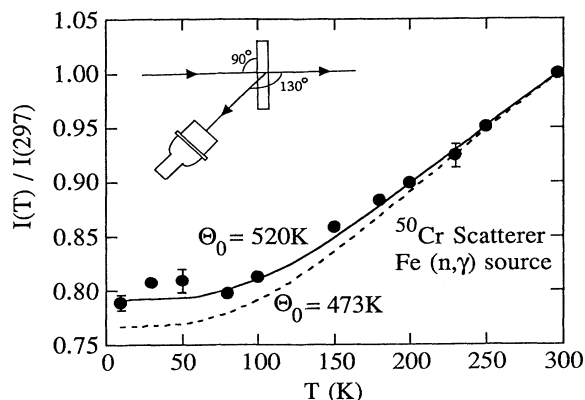


FIG. 5. Relative scattered intensities vs T from metallic Cr (relative to 297 K). The scatterer thickness was 5.3 g/cm^2 and the geometry is indicated in the figure. The solid line represents the best fit calculated curve obtained using $\delta=17 \text{ eV}$, $\Gamma=0.75 \text{ eV}$, $\Gamma_0/\Gamma=0.9$, and $\Theta_0=520 \text{ K}$ for the metallic Cr scatterer. The dashed line represents the calculated values for $\Theta_0=473 \text{ K}$. Some typical errors are indicated.

INS values were also obtained in previous NRPS measurements¹⁻⁴ and are listed in Table I.

The relatively large difference in Θ_0 obtained by the two methods as revealed in Table I may be partly explained by noting that the value of T_0 determined by NRPS monitors the vibrational frequencies of the Cr atoms at around 12 K (as explained above) while the phonon spectrum deduced from INS data is usually taken at 296 K. Thus a proper comparison can in principle be made only for a case where the phonon spectrum is known at 12 K. Some information about the behavior of the phonon spectra as a function of T is available in Ref. 11 for Li, Al, Cr, Rb, and Pd from which it is estimated that the phonon energies may increase by at most 3% when T decreases from 296 to 12 K. In the case of Cr, the error bars in the phonon spectrum may allow for another 2% increase in the phonon energies. It is thus possible to explain, at best, a 5% deviation between the NRPS and the INS methods. It is hard to attribute the remaining deviation solely to experimental errors because there is no reason why the NRPS measurements should always yield systematically higher results than that of the INS data. Another point worth noting is that different calculational methods (based on identical experimental data) may result in different phonon spectra. This point may be illustrated for the case of metallic Pb where

TABLE I. Metallic Debye temperatures for some isotopes determined by the present technique together with those obtained by integrating over the phonon spectra as in Eq. (7). The values of Θ_0 corrected for the composition of the natural elements are also listed.

| Isotope | NRPS | | Phonons ^a |
|-------------------|---------------------------------|---------------------------------|---------------------------------|
| | $\Theta_0(\text{K})$ isotope | $\Theta_0(\text{K})$ element | $\Theta_0(\text{K})$ element |
| ⁵⁰ Cr | 520 ± 20 | 510 ± 20 | 464 |
| ⁴⁸ Ti | 420 ± 20^b | 420 ± 20 | 360 |
| ⁶² Ni | 420 ± 15^c | 431 ± 15 | 384 |
| ⁶⁸ Zn | 235 ± 10^d | 240 ± 10 | 230 |
| ²⁰⁸ Pb | 104 ± 6^c | 104 ± 6 | 93 |

^a Reference 6. ^b Reference 3. ^c Reference 1. ^d Reference 2.

different calculations of $g(\nu)$ at 80 K carried out by different authors¹³⁻¹⁵ yielded values of Θ_0 ranging between 90 to 94 K. These values, though lower than our measured value (taken effectively at 12 K and listed in Table I) shows the differences in Θ_0 of $\approx 4\%$ results solely from the particular computational method used in deducing $g(\nu)$. It is therefore tempting to attribute this difference to some vibrational modes of the lattice which are monitored by the NRPS technique but not observed by INS. This is because with the present technique one can, at least in principle, view all the vibrations of the lattice [see Eq. (1)] while with INS from which the phonon spectra are deduced, it is not unlikely that some high-energy modes may be missed. This explanation of the deviation is qualitative and a detailed study of the phonon spectra at around 12 K is required to decide whether the above factor can account for all or only part of the discrepancy mentioned above.

In conclusion, this relatively large difference between the Debye temperatures deduced using the NRPS technique and that deduced from the phonon spectrum can only partially be explained; the source of the remaining difference of about 5% is unclear because at present we cannot identify any missing modes which are probably observed by the present technique but not with INS.

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