PHYSICAL REVIEW B VOLUME 33, NUMBER 7 1 APRIL 1986

Vibrational energies from nuclear γ resonant scattering: Measurement and comparison with neutron inelastic scattering

I. Jacob
Department of Nuclear Engineering, Ben-Gurion University of the Negev, 84105 Beer-Sheva, Israel

A. Wolf and O. Shahal
Department of Physics, Nuclear Research Centre, Negev, 84190 Beer-Sheva, Israel

R. Moreh*
Department of Physics, Nuclear Research Centre, Negev, 84190 Beer-Sheva, Israel
and Department of Physics, Ben-Gurion University of the Negev, 84104 Beer-Sheva, Israel

(Received 8 November 1985)

Nickel and lead lattice-vibrational energies (including the zero-point energies) are deduced at 298 and 80 K from nuclear γ resonant scattering measurements. A comparison is made with recently compiled neutron inelastic scattering data. The agreement is of the order of 1–2%. We conclude that the technique of nuclear resonant scattering of γ rays can be confidently used to obtain vibrational energies of specific atoms in various compounds. This type of information cannot presently be obtained from neutron inelastic scattering or any other experimental method.

Lattice vibrations and interatomic forces present one of the central problems in solid-state physics. Inelastic neutron scattering is presently the most powerful experimental tool for phonon investigations. An enormous amount of experimental and analytical work has been done on inelastic neutron scattering since the first phonon dispersion curves of Al were obtained in 1955.[1] The obvious need for tabulating the various results has recently been satisfied by a number of comprehensive compilations.[2–4] In this work we show that the technique of nuclear resonant scattering of gamma rays can also be used to obtain information on the lattice dynamics of various compounds. Specifically, the vibrational energies of particular atoms in the compound can be deduced. These energies are directly related to the phonon spectrum, and for pure metals, can also be computed from neutron inelastic scattering data. We will show that for two cases (i.e., nickel and lead), the agreement between the two methods: neutron inelastic scattering and resonant nuclear gamma scattering is very good. Thus, the latter technique can be used confidently to measure the vibrational energy of a specific atom (like Ni, Pb, and others) in a given compound. This type of selective information cannot be directly obtained from neutron inelastic scattering.

We consider the normalized frequency spectrum (density of phonon states as a function of frequency):

$$\int_0^\infty g(\nu)d\nu = 1 \ ,$$

where $g(\nu)d\nu$ is the number of phonon states per degree of freedom in the frequency range $(\nu, \nu+d\nu)$.

The mean energy per mode of vibration in the harmonic approximation is given by (e.g., Ref. 5)

$$\begin{aligned} \epsilon = k_B T_e = \int_0^\infty n(\nu,T)h\nu g(\nu)d\nu \ , \\ n(\nu,T) = [\exp(h\nu/k_B T) - 1]^{-1} + \tfrac{1}{2} \ . \end{aligned} \tag{1}$$

The mean energy, ϵ, includes also the zero-point energy. The notion of the effective temperature T_e was first introduced by Lamb.[6] He treated the thermal motion of the atoms in a solid in the same manner as the atoms of a gas, but using an effective temperature which is higher (or equal in a limiting case) than the thermodynamic temperature of the material. In the harmonic approximation, the mean vibrational kinetic (or potential) energy per vibrational mode equals exactly half of the corresponding total energy ϵ. The mean kinetic energy can be directly measured in a photon nuclear resonance scattering experiment. This experimental method utilizes an accidental overlap between an incident gamma line from some (n,γ) source and an excited nuclear level in a certain target nucleus. The overlap, represented schematically in Fig. 1, causes resonant scattering to occur. Excited nuclear levels at energies, $E_\gamma \cong 7$ MeV, usually display natural radiation widths Γ_0 between few meV (which is approximately the lowest detectable limit of the tech-

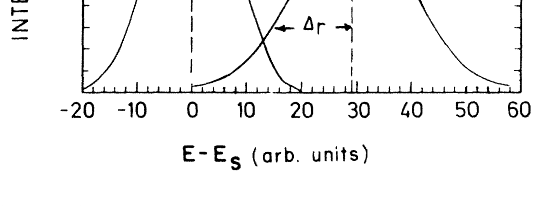

FIG. 1. The condition for resonant scattering of incident monoenergetic photons of peak energy E_r by an excited level of peak energy E_s. The two lines are Doppler broadened (Δ_r and Δ_s) and are separated by an energy δ (from Ref. 13).

nique) and 1 eV. The nuclear levels are however also Doppler broadened because of the thermal and zero-point vibrational motion of the scattering nuclei. The Doppler width Δ is given by

$$\Delta = E_\gamma \left[\frac{2kT_e}{Mc^2} \right]^{1/2} , \tag{2}$$

where M is the mass of the resonance nucleus and c is the velocity of light. The Doppler widths are of the order of 5–10 eV at room temperature for a wide range of atomic masses, M (50–200 amu), and are therefore the dominant factor determining the energy width of the resonant levels. The intensity of the scattered radiation depends on the relative position and overlap between the incident and the resonant gamma lines. In a more formal way, the scattered intensity $C(T,\theta)$ at angle θ and temperature T depends on the nuclear parameters Γ_0, Γ, δ, g of the level, on the effective temperature T_e of the resonant nucleus, and on the effective temperature T_s of the (n,γ) source:

$$C(T,\theta) = f(\Gamma, \Gamma_0, \delta, g, T_e, T_s) \ .$$

Γ and Γ_0 are the total and the partial ground-state radiative widths, respectively, δ is the energy separation between the incident γ line and the resonance level, and g is a statistical factor depending on the spins of the resonance and ground states. $C(T,\theta)$ is proportional to the resonance scattering cross section and its explicit form is given elsewhere.[7] Clearly, a change in the effective temperature affects the Doppler width [Eq. (2)]. Two ways for changing T_e will be mentioned. Stiffening of the phonon spectrum increases the mean total energy ϵ [Eq. (1)] and therefore increases the effective temperature and the Doppler width and vice versa; softening of the phonon spectrum decreases ϵ, T_e, and Δ. This can be achieved by forming different compounds of the same element. Another way to change T_e is by varying the temperature T of the scatterer, thus changing the occupation number $n(\nu,T)$—an increase of T increases ϵ, T_e, and Δ and vice versa. The mentioned change of the Doppler width may or may not cause a corresponding variation in the intensity of the resonantly scattered radiation. Such a variation depends on the specific relative position between the incident gamma line and the resonant level (Fig. 1), i.e., the resulting overlap may or may not be influenced by the change in Δ. A temperature variation experiment may serve as a convenient probe for the sensitivity of the scattered intensity in a certain source-target combination to a change of the atomic kinetic energy (i.e., T_e or Δ). More specifically, it is convenient to utilize as a probe the temperature effect, R, defined as the ratio

$$R(T_1, T_2, \theta) = \frac{C(T_1,\theta)}{C(T_2,\theta)} \ . \tag{3}$$

A convenient set of temperatures T_1, T_2 is $T_1 = 77$ K (liquid nitrogen) and $T_2 = 300$ K (room temperature). A suitable (n,γ) source-scatterer combination for investigating the mean vibrational kinetic energy should provide a relatively high scattered intensity and a significant temperature effect ($R \leq 0.95$ for the above T_1, T_2).

Table I lists some of the intense scatterers together with the corresponding energies and *measured* R values. From the measured R values T_e may be calculated at one of the temperatures T_i $(i = 1,2)$, provided T_e at the second temperature is known. However, usually one cannot assume this to be the case, and a determination of $T_e(T)$ requires a careful measurement of $C(T,\theta)$ over a wide temperature range including high temperatures ($T >> \theta_D$, the Debye temperature). At high temperatures, one can safely assume that $T_e \cong T$ and consequently calculate $T_e(T)$ at lower temperatures. Such a resonant scattering experiment over a wide temperature range has been performed only for two elemental metals, namely, Ni and Pb.[10] It should be noted in this context that an extensive research work on the binding properties of nitrogen in molecules has been carried out utilizing the present technique (e.g., Refs. 11–13 and references therein). Although the information of $T_e(T)$ for Ni and Pb is inherently contained in the data presented in Ref. 10, explicit values of $T_e(T)$ were not deduced there. It is of special interest to compare the T_e values from the nuclear resonant scattering experiment with those available from

TABLE I. Some resonant scatterers with significant temperature effect R (T_1, T_2, 135°) (from Refs. 8 and 9).

(n,γ) source	Resonant scattering isotope	Percent abundance	Resonance energy (MeV)	Temperature effect R[a]
Fe	^{62}Ni	3.66	7.646	0.90 ± 0.006
Fe	^{208}Pb	52.3	7.279	0.937 ± 0.003
Fe	^{50}Cr	4.3	8.888	0.84 ± 0.02
Fe	^{141}Pr	100	7.632	0.95 ± 0.01
Fe	^{139}La	99.9	6.018	0.969 ± 0.003
Ni	^{86}Sr	9.9	7.82	0.96 ± 0.01
Ni	^{150}Sm	7.4	8.998	0.95 ± 0.007
V	^{48}Ti	74	6.600	0.80 ± 0.02
V	^{146}Nd	17.2	7.163	0.93 ± 0.01
Cr	^{15}N$_2$	0.366	6.324	0.72 ± 0.03
Cr	^{68}Zn	18.6	7.362	0.773 ± 0.01[b]
Ti	^{65}Cu	30.9	6.556	0.94 ± 0.02

[a] The temperature effect R for N_2 was measured at $T_1 = 115$ K, $T_2 = 293$ K and at $T_1 = 77$ K, $T_2 = 300$ K for the rest of the resonant scatterers in the table.

[b] R. Moreh, O. Shahal, and J. Tenenbaum, J. Phys. G **9**, 755 (1985).

TABLE II. Effective temperatures T_e, in Kelvin, of Pb and Ni deduced from inelastic scattering data and nuclear resonant gamma scattering experiment, at temperatures T, for which the former information is available.

	Neutron inelastic scattering		Resonant gamma scattering	
T (K)	Ni (Ref. 14)	Pb (Ref. 15)	Ni	Pb
80		86.2		87 ± 1
298	321.9		$327.4 \pm 3 (324.5 \pm 3)$[a]	
676	684.9		691 ± 1[b]	

[a]This value is deduced after adopting the effective temperature of 684.9 K for Ni at 676 K from the neutron inelastic scattering.

[b]This is an assumed value of T_e which is very close (2%) to the actual thermodynamic temperature (see also text).

neutron inelastic scattering. In the last case the derivation is straightforward if the actual form of $g(\nu)$ and Eq. (1) are utilized. Such a comparison is much easier to make today with the aid of one of the excellent and comprehensive compilations mentioned above.[3] The comparison is presented in Table II at temperatures for which the $g(\nu)$ functions are tabulated in Ref. 3. The agreement is very good—it is within 1% for lead and slightly worse (1.5%) for nickel. In Pb the ratio between the maximum temperature, at which the resonance scattering was measured, and the cut-off frequency of the phonon spectrum significantly exceeds the corresponding ratio in Ni. We employ for convenience the concept of the Debye temperature, θ_D, as a measure of the cut-off frequency. The mentioned maximum temperature for Pb is 520 K and it should be compared with $\theta_D = 104$ K, while in Ni the corresponding values are 670 K and $\theta_D = 420$ K.[10] This situation may explain the slightly better agreement with the neutron inelastic scattering results because in the specific case of Pb the high-temperature assumption $T_e = T$ is a better approximation for Pb than for Ni. We have assumed (see Table II) that $T_e(\text{Ni}, T = 676 \text{ K}) = 691$ K. This value is higher by about 2% and 1% than the actual temperature and the corresponding "neutron" T_e, respectively. Adopting the latter T_e for deducing the effective temperature of Ni at 298 K by our method further improves the agreement with the neutron inelastic scattering result (Table II). In Fig. 2, the temperature effect in Pb is shown versus T_e in order to demonstrate the method of analyzing our results. The sloped line in Fig. 2 exhibits the calculated temperature effect $R(540 \text{ K}, T, 135°)$ as a function of various arbitrary T_e values at temperature T. $T_e(T = 540 \text{ K})$ is assumed to be known and equal to 540 K. The horizontal lines indicate the experimental values of $R(540 \text{ K}, 80 \text{ K}, 135°)$ and the respective errors. The intersection of the horizontal lines with the calculated curve determines the T_e value of Pb at 80 K. It is clearly seen that the experimental accuracy decreases with increasing of T_e at a given thermodynamic temperature T. However for these large values of T_e, the significant deviation of T_e from T compensates for the lower accuracy.

It should be stressed that in the case of pure elements, the neutron inelastic scattering provides much more comprehensive and detailed information than the gamma resonant scattering experiment. The importance of the

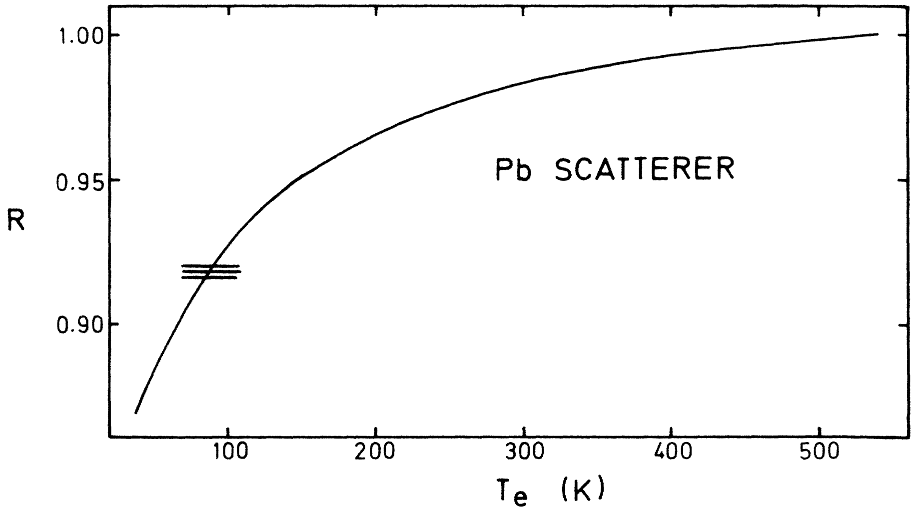

FIG. 2. Calculated values of the temperature effect $R(540 \text{ K}, T, 135°)$ [defined in relation (3)] of elemental Pb with a fixed $T_e(T = 540 \text{ K}) = 540$ K vs T_e. The horizontal lines indicate the experimental value of $R(540 \text{ K}, 80 \text{ K}, 135°)$ and the respective errors. The scatterer thickness, the geometry, and the nuclear parameters are given in Ref. 10.

above comparison is to show the validity of the photon nuclear resonance technique for measuring vibrational energies. Our results may be of great value in cases (e.g., intermetallics, hydrides, carbides, nitrides, and other compounds) where such information is additional to that provided by the neutron inelastic scattering. The mean energy per vibrational mode of the ith component ϵ_i in a compound of n components will be given in terms of its effective temperature, $T_{i,e}$ by

$$\epsilon_i = k_B T_{i,e} = \frac{\int_0^\infty S_i(\nu)\, n(T,\nu)\, h\nu\, g(\nu)\, d\nu}{N_i/N} ,$$

where $S_i(\nu)$ is the fraction of the phonon energy carried by the ith atom at frequency ν. N_i is the number of the i atoms in the sample and N is the total number of atoms in it. Clearly, $\sum_{i=1}^{n} N_i = N$ and N_i/N is the fraction of the degrees of freedom belonging to the i atoms.

The derivation of $S_i(\nu)$ for a certain component in a compound is a complicated question and is associated with the determination of the eigenvectors for a certain set of eigenfrequencies. In other words, the quantity ϵ_i is not directly accessible from the neutron inelastic scattering experiment and it is model dependent. On the other hand, the nuclear resonant scattering of gamma rays can provide ϵ_i of certain elements directly and independent of any model. The mean kinetic energy per vibrational mode from the gamma resonant scattering may be regarded as being independent, even of the harmonic approximation which is assumed only in order to compare with the results of the inelastic neutron scattering. We propose two experimental ways for deducing $T_e(T)$ of certain elements (some of them are presented in Table I) either in their elemental form or as a component of a compound. The first one requires a measurement of the resonantly scattered radiation over a wide temperature range up to high enough T values, as was done for Ni and Pb. In the second, information for T_e at a given temperature for a specific element may be obtained from inelastic neutron scattering data. Then T_e of this element in any compound and at any temperature may be deduced by comparing the corresponding intensity of the gamma resonant radiation with the scattered intensity of the pure element at the given temperature. Both ways present some experimental difficulties which we believe may be overcome.

The values of ϵ_i obtained from nuclear resonant scattering experiments, can be of great value in the interpretation of lattice dynamics calculations, in which different models yield different values of ϵ_i. Comparison of these values with the experimental results can help to differentiate between the various models.

In summary, very good experimental agreement with regard to vibrational energies was shown to exist between neutron inelastic scattering experiments and the nuclear gamma resonant scattering method. This agreement proves the validity of using the latter technique for measuring vibrational energies of specific elements, either in pure form or in compounds. Some future directions of research (mainly with compounds) have been proposed and discussed.

The authors wish to thank J. Ashkenazi for stimulating talks.

---

[*] Present address: Physics Department, Brookhaven National Laboratory, Upton, NY 11973.

[1] B. N. Brockhouse and A. T. Stewart, Phys. Rev. **100**, 756 (1955).

[2] H. Bilz and W. Kress, *Phonon Dispersion Relations in Insulators* (Springer, Berlin, 1979).

[3] H. Schober and P. H. Dederichs, in *Phonon States of Elements, Electron States and Fermi Surfaces of Alloys*, Landolt-Bornstein, New Series, Group III, Vol. 13a, edited by K. H. Hellwege and K. H. Olsen (Springer, Berlin, 1981).

[4] W. Kress, in *Phonon States of Alloys, Electron States and Fermi Surfaces of Strained Elements*, Landolt-Bornstein, New Series, Group III, Vol. 13b, edited by K. H. Hellwege and K. H. Olsen (Springer, Berlin, 1983).

[5] C. Kittel, *Introduction to Solid State Physics* (Wiley, New York, 1971), p. 205.

[6] W. E. Lamb, Jr., Phys. Rev. **55**, 190 (1939).

[7] R. Moreh, S. Shlomo, and A. Wolf, Phys. Rev. C **2**, 1144 (1970).

[8] A. Wolf, Ph.D. thesis (in Hebrew), Weitzman Institute, Rehovot, 1973, also Nuclear Research Center-Negev Report No. 343 (unpublished).

[9] R. Moreh and O. Shahal, Nucl. Phys. **A252**, 429 (1975).

[10] R. Moreh, O. Shahal, and I. Jacob, Nucl. Phys. **A228**, 77 (1974).

[11] O. Shahal, R. Moreh, and M. Pazi, Nucl. Phys. **A339**, 157 (1980).

[12] R. Moreh, W. S. Sellyey, and R. Vodhanel, Phys. Rev. C **22**, 1820 (1980).

[13] R. Moreh, Nucl. Instrum. Methods **166**, 45 (1979).

[14] G. A. de Wit and B. N. Brockhouse, J. Appl. Phys. **39**, 451 (1968), the data are presented in Ref. 3, p. 104.

[15] R. Stedman, L. Almquist, and G. Nillson, Phys. Rev. **162**, 549 (1962); E. R. Cowley, Solid State Commun. **14**, 587 (1974), the data are presented in Ref. 3, p. 108.