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Study of Atomic Velocities in Molecules Using Nuclear Resonance Photon Scattering

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A strong dependence of the scattering cross section on molecular orientation was observed when 6.324-MeV photons were scattered from ^{15}N in the form of $\mathrm{Na^{15}NO_3}$ single crystal. This is caused by the different velocities of the N atom corresponding to the various directions of the *zero-point* molecular vibrations.

We have measured the resonance scattering cross section of 6.324-MeV photons[1,2] from a ^{15}N target in the form of a $\mathrm{Na^{15}NO_3}$ single crystal. A strong dependence of the scattering cross section on crystal orientation was observed. This behavior is due to the anisotropic motion of the N atom in the crystal which causes different Doppler broadenings of the nuclear level in the variour directions. The results are used to determine the velocity component of the N atom along the photon beam direction. The most important point to be noted here is that the dominant part of this velocity is due to the *zero-point vibration* of the N atom in the various normal modes of vibration of the molecule. The excited vibrational states do not contribute in practice as they are almost unpopulated at room temperature because of the high vibrational energies involved.

The 6.324-MeV level of ^{15}N is photoexcited by a chance overlap (to within ≈ 30 eV) of one of the incident γ lines of the reaction $\mathrm{Cr}(n, \gamma)$. The technique involved is described in detail elsewhere.[3]

The Doppler broadening of both the incident γ line and the resonance level in ^{15}N causes an appreciable overlap between the tails of the two lines which gives rise to a relatively high scattering cross section σ_s. σ_s is very sensitive to the Doppler width of the resonance level and hence to the instantaneous velocity of the N atom. In addition, the lifetime of the resonance level ($\tau \sim 10^{-16}$ sec) is much shorter than the period of the molecular vibrational motion ($t \leqslant 10^{-14}$ sec), and hence the photon is emitted long before any "smearing" of the vibrational velocity takes place. For nuclear levels where $\tau \gg t$, the zero-point vibration can cause almost no Doppler broadening and the effect reported here cannot be observed.

We now consider the motion of the N atom in $\mathrm{NaNO_3}$. The $\mathrm{NO_3^-}$ molecular ions are planar[4] with their plane perpendicular to the hexagonal *c* axis of the $\mathrm{NaNO_5}$ unit cell. Because of the weak ionic binding between $\mathrm{Na^+}$ and $\mathrm{NO_3^-}$, the kinetic energy of the N atom is due primarily to its internal vibrational motion in $\mathrm{NO_3^-}$. In its ground state, the $\mathrm{NO_3^-}$ molecular ion performs zero-point vibrations corresponding to six normal-mode frequencies.[2,4] The first mode does not contribute to the motion of the N atom. In the second mode, ν_2, the N atom vibrates along the

In the remaining four modes (ν_j, $j=3,\ldots,6$), the N motion is restricted to the NO_3^- plane and the velocity component of the N atom is the same for all directions within the plane. This follows from the symmetry of the NO_3^- ion. The Doppler widths characterizing these modes of the zero-point directional vibrations of the N atom can be entirely different and can be detected by mounting a $Na^{15}NO_3$ single crystal so that its *c* axis is parallel to the photon beam ($\theta=0$). Thus, the measured scattered photon intensity as a function of θ varies smoothly according to the Doppler broadening caused by the velocity components of the normal modes of vibration.

Experimentally, the photon beam was obtained from the (n,γ) reaction on some chromium disks placed along a tangential beam tube[3] and near the core of the IRR-2 reactor. The photon beam contains among several other γ lines, a line at 6.324 MeV whose intensity is $\sim 10^6$ photons/cm^2 sec at the target position. The target consisted of a 200-mg $Na^{15}NO_3$ single crystal (99.7% ^{15}N) mounted in such a way that its *a* axis coincided with the axis of a stepping motor used for rotating the crystal. An automatic control system was employed for measuring the number of scattered photons as a function of θ. The scattered radiation was detected using a 12.7×12.7-cm^2 NaI crystal placed at 120° with respect to the incident beam, with its front face 18 cm away from the target. The number of counts versus θ was recorded directly on a multichannel analyzer operating in a multiscaler mode. The result given in Fig. 1 shows that σ_s varies from a maximum value σ_a for which the incident photon direction lies in the NO_3^- plane ($\theta=90°$) to a minimum value σ_s for the perpendicular direction ($\theta=0°$). Experimentally we found that $\sigma_a/\sigma_c=1.43\pm0.03$. By combining this ratio with the measured scattering cross section for an amorphous $Na^{15}NO_3$ sample ($\sigma_e=2.48\pm0.13$ b) and by using the relation

$$\sigma_e=(\sigma_c+2\sigma_a)/3 \qquad (1)$$

(where the factor 2 allows for the two degrees of freedom of the planar motion of the N atom in the NO_3^- plane), we obtain $\sigma_c=1.93\pm0.10$ b and $\sigma_a=2.76\pm0.14$ b.

These cross sections correspond to different Doppler widths of the 6.324-MeV level in ^{15}N and are caused by different effective temperatures T_c and T_a of the N atom along the *c* axis and the *a* axis of the NO_3^- molecule. From the values of σ_c and σ_a, and using Fig. 2, we find that $T_c=(575\pm25)$°K and $T_a=(843\pm40)$°K, for a crystal temperature of $T=293$°K.

Figure 2 gives a plot of the calculated values of σ_s (for the 6.324-MeV resonance level in ^{15}N) versus the effective temperature T_e of the N atom, where

$$\sigma_s=\sigma_0^m\psi(x,t), \qquad (2)$$

$\sigma_0^m=h^2c^2(2J+1)/(2J_0+1)2\pi E_0^2$, with J_0 and J the respective spins of the ground and resonance levels of ^{15}N. The function ψ was defined in Refs. 2 and 3.

It should be noted that T_c and T_a give a measure of the velocity and the *total* kinetic energy E_k of the N atom along the two perpendicular directions and were first defined by Moreh, Shahal, and Volterra[2] who generalized the treatments of

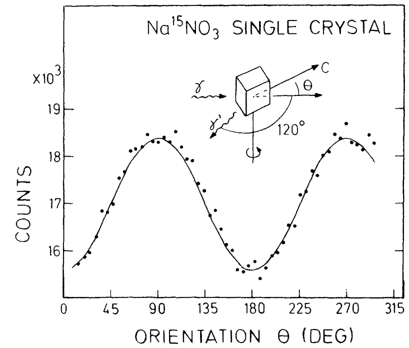

FIG. 1. Number of counts versus angular orientation θ obtained by rotating the 200-mg $Na^{15}NO_3$ single crystal (99.7% ^{15}N) in angular steps of $\Delta\theta=5.4°$. A 12.7×12.7-cm^2 NaI detector was used. The total running time was ~2 days. The solid line is a plot of $\sigma_s=\sigma_c+(\sigma_a-\sigma_c)\sin^2\theta$.

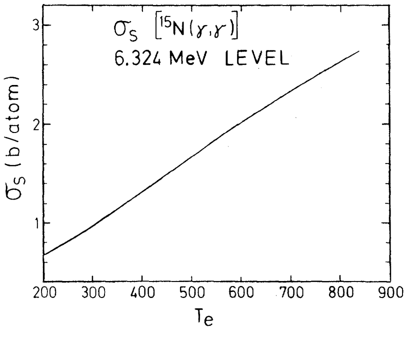

FIG. 2. Calculated scattering cross section [Eq. (2)] of the 6.324-MeV γ [from the reaction Cr(n,γ)] from ^{15}N as a function of the effective temperature T_e of the N atom. The parameters of the resonance level were taken from Ref. 2.

NO_3^- c axis. Lamb[5] and Metzger[6] to the case of complicated molecules.

Note that T_c and T_a can also be expressed theoretically by using results of Ref. 2 as follows:

$$T_c = \frac{M_0}{M_M} T_r + S_2 \frac{h\nu_2}{k}\left(\frac{1}{\exp(h\nu_2/kT)-1} + \frac{1}{2}\right), \quad (3)$$

$$T_a = \frac{M_0}{M_M} T_r + \sum_{j=3}^{6} \frac{h\nu_j}{2k}\left(\frac{1}{\exp(h\nu_j/kT)-1} + \frac{1}{2}\right), \quad (4)$$

where $M_0/M_m = 15/63$ is the mass ratio of ^{15}N to the $^{15}NO_3^-$ ion T_r is an effective temperature obtained by applying the Lamb treatment to the whole NO_3^- molecule and was described in more detail earlier.[2] Here we assume that T_r is independent of the directional properties of the NO_3^- molecule. S_j is the fraction of the kinetic energy shared by the N atom in NO_3^-, and ν_j ($j = 2, \ldots, 6$) refers to the normal-mode frequencies.[4] The values of S_j may be obtained by solving the equations of motion of the NO_3^- molecule.[2]

By using Eqs. (3) and (4) and the values of S_j and ν_j from Ref. 2, we obtain $\Delta T = T_a - T_c = 251°\mathrm{K}$, to be compared with a measured value $\Delta T = 268°\mathrm{K}$ found experimentally here. The deviation is probably due to the anisotropic motion of the NO_3^- ion in the single crystal. For an amorphous sample, the effective temperature is given, in analogy with Eq. (1), by

$$T_e = (T_c + 2T_a)/3. \quad (5)$$

In order to explain the behavior of σ_s versus θ (Fig. 1), we note that the effective temperature along the θ direction depends on the average sum of the kinetic energies of the N atom along this direction; hence

$$T_e(\theta) = T_c + (T_a - T_c)\sin^2\theta \quad (6)$$

and the corresponding scattering cross section σ_s is therefore

$$\sigma_s(\theta) = \sigma_c + (\sigma_a - \sigma_c)\sin^2\theta. \quad (7)$$

Equation (7) follows by noting that σ_s varies linearly with T_e (Fig. 2). It yields an excellent fit to the experimental data (Fig. 1).

In conclusion, the present measurement establishes, in effect, a new temperature scale which treats the internal kinetic energy of the zero-point vibrational motion (of the N atom in a molecule) on the same footing as that of the translational thermal motion. Alternatively, it can also be used for measuring the velocity component of the zero-point vibrational motion of an atom in a molecule.

The present technique illustrates the possibility of varying the temperature of the N atoms by simply varying the molecular orientation or by using different N-containing molecules. This technique may also prove useful for studying the orientation of adsorbed molecules on ordered substrates in cases where it is otherwise impossible to study with other methods.

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