Inelastic-Scattering Measurements of 1.5–15 eV Neutrons

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Measurements of inelastically scattered electronvolt neutrons have been completed with a pulsed neutron source and neutron time-of-flight techniques in combination with a resonant-neutron-capture detector. Measurements are presented on liquid nitrogen and benzene for incident neutron energies in the range 1.5 to 15 eV and for q values from 13 to 120 Å^{-1}. These are the first measurements of inelastic neutron scattering in this energy range.

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Studies of matter using inelastically scattered neutrons have been restricted to excitation energies below about 0.3 eV primarily because the neutron spectrum from a research reactor decreases rapidly above 0.1 eV and also because reactor experimental methods are not easily extended to higher energies. With the increasing availability of pulsed spallation sources, which are excellent sources for neutrons in the electronvolt range, there is growing interest in developing experimental methods for electronvolt inelastic scattering. Such research offers the possibility of studying high-lying molecular rotational-vibrational states, molecular electronic excitations, and electronic levels in solids which can only be studied by using electromagnetic probes and even with these often not in bulk material. In this Letter we report the first measurements of inelastic neutron scattering in the electronvolt region.

Two types of neutron interactions appear to be promising; excitation arising from magnetic interactions between the neutron and electron, which will not be considered further here, and excitation by hard-sphere neutron-nucleus scattering. The experimental problem in these studies is the simultaneous measurement of both the incident and scattered neutron energies with the time-of-flight method. Of the several possible solutions, the approach chosen here is the detection of the scattered neutrons by measuring capture γ rays from an absorber with a strong isolated electronvolt resonance. This method is ideally suited for measurements using a pulsed white source of neutrons.

The geometry for our experiment is shown in Fig. 1. The source of neutrons is the National Bureau of Standards 100-MeV electron linac using a tungsten target and operating with a pulse width of 2 μs, an average power of 3.5 kW, and a pulse rate of 360 Hz. The total average 4π neutron production rate is about 7×10^{12} n/sec. The neutrons are moderated in CH_2 and separated in energy by time-of-flight measurements along a 21-m drift tube. The beam is collimated at the sample to a 5×10 cm^2 rectangle. Scattered neutrons travel about 15 cm to an In or Au foil placed against the surface of a C_6D_6 liquid scintillator surrounded on five sides with lead shielding. By measuring the flight time from source to scatterer to detector, the incident energy can be determined since the final energy is fixed by the resonance. The resolution ultimately is limited by the full width at half maximum of the gold
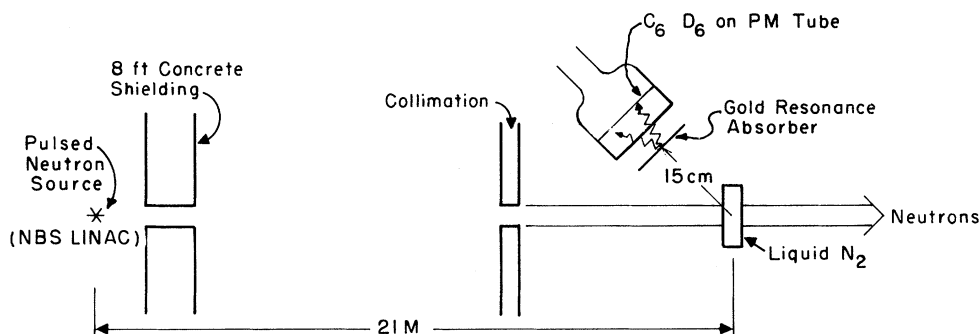

FIG. 1. Arrangement for detecting electronvolt inelastic neutron scattering by the time-of-flight method.
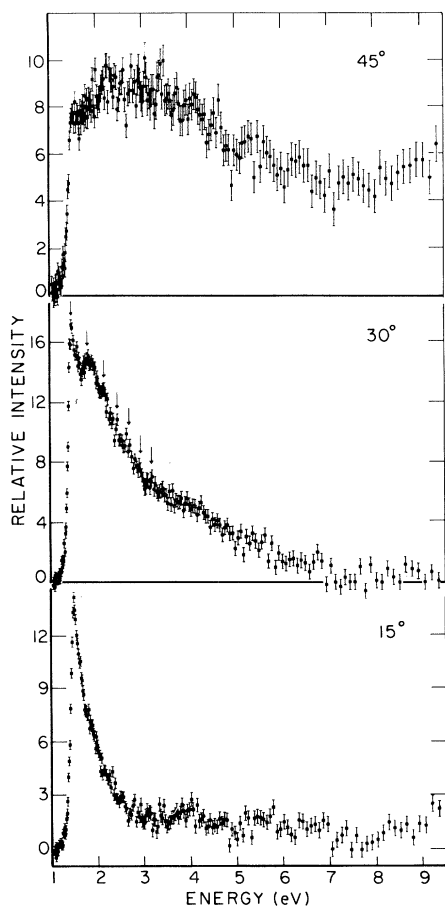
FIG. 2. The spectrum of inelastically scattered neutrons from a benzene sample. The neutron detection rate is plotted vs the incoming neutron energy. The final neutron energy is fixed by the indium resonance to be 1.46 eV. The difference between the incoming energy and the final energy is the excitation energy of the molecule.
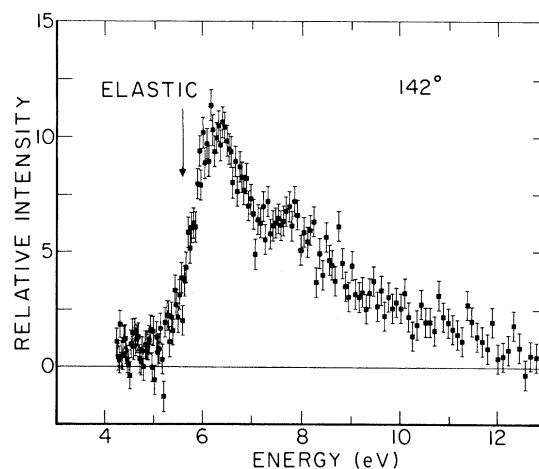

FIG. 3. The spectrum of inelastically scattered neutrons from a liquid nitrogen sample using an Au absorber with resonance at 4.9 eV. The peak at 7.8 eV arises from an uncorrected background.

and indium resonances which are 72 and 140 meV, respectively. Backgrounds are significantly reduced by using the organic scintillator loaded with deuterium to eliminate bothersome γ rays from capture of moderated neutrons in hydrogen. Most of the background can be measured by absorber-in, absorber-out comparison.

The results of our experiments on a 0.5-cm-thick sample of liquid benzene using an indium resonance absorber with a resonance at 1.46 eV are given in Fig. 2. Spectra are shown for scattering angles of 15°, 30°, and 45°. The incident neutron energy is shown on the abscissa and the neutron detection rate in the indium resonance per timing channel is given on the ordinate. The excitation energy of the molecule is the difference between the incident energy on the abscissa

and 1.46 eV. The increased probability of higher molecular excitation energy with increasing scattering angle is clearly evident. For the 30° spectrum the momentum transfer varies from $q = 10$ to $q = 50$ Å^{-1}. For this spectrum, which was measured for the longest time (24 h), the statistical accuracy is sufficient to clearly show structure in the scattered spectrum. The structure might arise from carbon-hydrogen stretching vibrations, which are commonly found by other methods in many organic molecules at about 0.37 eV.

The 45° spectrum suggests that electronic states are perhaps being excited. For 9.5-eV incoming neutrons, the energy deposited in the molecule is about 8 eV. This energy may be compared with 5 eV lost when the neutron scatters at 45° from a free hydrogen atom. The lowest-lying electronic state in benzene is at 3.7 eV.[1] Therefore below $1.45 + 3.7 = 5.2$ eV the excitation energy is carried entirely by a combination of vibrational modes. The moment of inertia of the molecule is too large for much energy to be associated with rotations. Measurements which are now possible with a shorter flight path, better shielding, and improved background subtraction should reveal any structure present within the resolution limitation of the indium resonance.

Figure 3 shows measurements carried out with a 1-cm-thick sample of liquid nitrogen using a gold resonance-capture absorber with a resonance at 4.9 eV. For neutrons in the 5–15 eV range and a scattering angle of 140°, q ranges

from 90 to 120 Å^{-1}. The vertical line at 5.7 eV marks the threshold for elastic scattering which is displaced from the gold resonance energy of 4.9 eV by center-of-mass recoil of the nitrogen molecule. The scattering measured below 5.7 eV arises from Doppler spreading, resolution, and neutron scattering with energy gain from the molecule. The curve peaks at 6.2 eV and has a broad tail which was measured up to about the threshold for electron excitations which lies approximately at an energy for incident neutrons of about 12 eV.[2] Thus the molecular excitation in Fig. 3 consists solely of rotational-vibrational excitations. The bump at 7.5 eV is associated with a small amount of antimony in the lead shield around the detector. For both the nitrogen and the benzene, the neutron intensity is approximately proportional to $E^{1/2}$. Therefore, both the benzene and nitrogen data can be transformed to a

quantity proportional to cross section by dividing the measured spectrum by $E^{1/2}$.

No neutron inelastic-scattering measurements have previously been made in the electronvolt energy range, and little work has been published on the probabilities for various modes of molecular excitation (in this energy range). However, Letokhov and Minogin have published excitation probabilities for diatomic[3] and centrosymmetric[4] molecules excited by radioactive γ decay of an isomeric nuclear state. These results can be carried over to the case of electronvolt inelastic neutron scattering, since the mode of excitation, i.e., recoil of one of the nuclei in the molecule, is exactly the same.

For a diatomic molecule with $M_1 = M_2$, the probability $P(V_b, V_a)$ of exciting a final vibrational state V_b from the initial state V_a when the momentum transfer is directed along the axis is given by the distribution

$$P(V_b, V_a) = \frac{V_a!}{V_b!} Z_0^{V_b - V_a} \exp(-Z_0)[L_{V_a}^{V_b - V_a}(Z_0)]^2,$$ (1)

where $Z_0 = q^2 \hbar^2 / M \hbar \omega$. The quantity q is the momentum transfer, $\hbar \omega$ is the characteristic energy of the harmonic oscillator, M is the reduced molecular mass, and L is the Laguerre polynomial. For the case of $V_a = 0$, (1) reduces to a Poisson distribution.

For a rigid rotor the probability $P(J_b, J_a)$ of a transition from the ground state J_a to the state J_b is found[3] to be

$$P(J_b, J_a) = (2J_b + 1) \sum_{l = J_a - J_b}^{J_a + J_b} (2l + 1) j_l^2(a_0) \begin{pmatrix} J_a & J_b & l \\ 0 & 0 & 0 \end{pmatrix}^2.$$ (2)

The quantity $a_0 = \hbar q / 2R_0$. The parameter R_0 is the equilibrium separation between the nuclei, and j_l is the half-integer Bessel function. For the case of $J_a = 0$, (2) reduces to the form

$$P(J_b, 0) = (2J_b + 1) j_{J_b}^2(a_0).$$ (3)

In general when both vibrational and rotational transitions are involved, the solution cannot be expressed in common mathematical functions. Thus in order to fit the nitrogen spectrum shown in Fig. 3 a numerical solution, which is beyond the scope of the present Letter, would be required. However, it is of interest to compare experiment with the theory based on vibrations alone. The usefulness of this comparison depends on the assumption that the average internal energy imparted to the molecule is the same regardless of the angle between the molecular axis and the momentum transfer vector. For nitrogen, $\hbar \omega = 0.289$ eV and for this data $Z_0 \simeq 2.5$ (slowly increasing). Applying Eq. (1) (for the case $V_a = 0$) we find that the peak of the distribution is at 6.2 eV and that it falls to half maximum at ~ 6.9 eV.

Both of these predictions are in approximate agreement with the observed spectrum, but a detailed comparison with experiment must include rotations.

Obviously, for the more complicated molecule benzene, these theoretical results, which were derived for a diatomic molecule, cannot be applied directly. In principle the theory can be extended and in this case where the scattering is primarily off hydrogen perhaps consideration of only a few normal modes will be sufficient.

Electronic excitations can be induced by neutron interactions through a direct magnetic interaction (which will not be considered here) or through the disturbance of the molecule resulting from the neutron collision with one of the nuclei of the molecule. This collision produces a recoil of the molecule as a whole and will also in general excite internal degrees of freedom. Letokhov and Minogin[3] have calculated the probability of an electron excitation due to the sudden acceleration of the molecular center of mass. The matrix

elements required are simply the overlap of the wave function of the molecule initially at rest with the wave function of the molecule accelerated to some final velocity. We have extended this work somewhat to calculate the probability for an homonuclear diatomic molecule to remain in its electronic ground state following a neutron interaction. This probability is

$$P_{oe} = 1 - \left(\frac{m_e}{M}\right)^2 \left\{ \langle \psi_{oe} | (\vec{q} \cdot \sum_i \vec{r}_i)^2 | \psi_{oe} \rangle - |\langle \psi_{oe} | \vec{q} \cdot \sum_i \vec{r}_i | \psi_{oe} \rangle|^2 \right\} , \tag{4}$$

where ψ_{oe} is the electronic wave function for the ground state, m is the mass of one of the nuclei, $\vec{r}_i$ is the distance from the center of mass to the ith electron, and m_e is the electron mass. The second term of (4) is therefore the probability for an electron excitation. Because of the $(m_e/M)^2$ term the electronic excitation probability will be small ($\sim 10^{-4}$) for inelastic scattering of electronvolt neutrons. However, it should be noted that electronic excitation accompanying neutron nuclear scattering should be the rule for kiloelectronvolt neutrons.

An additional mode of electronic excitation can arise from nonadiabatic effects in molecular physics. In the usual (adiabatic) approximation the molecular wave function is separable[5] into an electronic component and components describing the internal degrees of motion of the nuclei, i.e., vibrations and rotations. This separation is satisfactory as long as the "nonadiabatic" terms in the full Schrödinger equation can be neglected. Born and Oppenheimer[6] showed that these terms may be safely neglected for the energy-level calculations although not necessarily for transition-probability estimates. These nonadiabatic terms can be included as a perturbation to the electronic wave functions but the necessary calculation is beyond the scope of this Letter. Preliminary work on such a calculation has been completed and will form part of a later publication.[7]

In summary, it appears that vibrational, rotational, and perhaps electronic transitions can be excited with measurable probability through electronvolt neutron-nucleus collisions. An experimental method with many possibilities for extension has been demonstrated. The basis, therefore, appears to be established for a general study of excitations by inelastic-scattering measurements with electronvolt neutrons.

[1]R. S. Mulliken, J. Chem. Phys. 46, 675 (1949).

[2]The first electronic state in N_2 lies at 6.1 eV. See G. Herzberg, Spectra of Diatomic Molecules (Van Nostrand Reinhold, New York, 1950), p. 449.

[3]V. S. Letokhov and V. G. Minogin, Zh. Eksp. Teor. Fiz. 42, 1569 (1975) [Sov. Phys. JETP 42, 800 (1975)].

[4]V. S. Letokhov, Phys. Rev. A 12, 1954 (1975).

[5]See, for example, G. Herzberg, Spectra of Diatomic Molecules (Van Nostrand Reinhold, New York, 1950), p. 148.

[6]M. Born and R. Oppenheimer, Ann. Phys. (N.Y.) 84, 457 (1927).

[7]S. W. Lovesey, C. D. Bowman, and R. G. Johnson, to be published.