Conf-820942--8
CONF-820942--8
DE82 022342

SOLID-STATE EFFECTS ON THERMAL-NEUTRON CROSS SECTIONS AND ON LOW-ENERGY RESONANCES

J. A. Harvey, H. A. Hook, N. W. Hill and O. Shahal†
Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830 USA

The neutron total cross sections of several single crystals (Si, Cu, sapphire), several polycrystalline samples (Cu, Fe, Be, C, Bi, Ta), and a fine powder copper sample have been measured from 0.002 to 5 eV. The Cu powder and polycrystalline Fe, Be and C data exhibit the expected abrupt changes in cross section. The cross section of the single crystal of Si is smooth with only small broad fluctuations. The data on two "single" Cu crystals, the sapphire crystal, cast Bi, and rolled samples of Ta and Cu have many narrow peaks ~ 10^{-3} eV wide. High resolution (0.3%) transmission measurements were made on the 1.057-eV resonance in ^{240}Pu and the 0.433-eV resonance in ^{180}Ta, both at room and low temperatures to study the effects of crystal binding. Although the changes in Doppler broadening with temperature were apparent, no asymmetries due to a recoilless contribution were observed.

[Si, Cu, Be, C, Bi, Ta, Sapphire, σ_T, E_n = 0.002-5 eV, 1.057-eV resonance ^{240}Pu, solid state effects, Doppler broadening]

Introduction

In the thermal energy region the measured total cross section often depends on the physical form of the sample. The total cross sections of polycrystalline materials such as Be, C and Fe show abrupt changes of ~ 2 barns or ~ 30% at energies corresponding to scattering at 180° from lattice planes in the crystal as shown in Figure 1. At higher energies, $\gtrsim$ 0.2 eV, so many planes are involved that the cross section is quite smooth (providing there are no nuclear resonances). At energies $\gtrsim$1 eV, the recoil energy is sufficient that the atoms are knocked out of the lattice and the free atom cross section is observed. Severe self-shielding or extinction effects can occur in the thermal energy region if the crystal grains are not very fine (~ 10^{-4} cm); the observed cross section of a polycrystalline material can be reduced by several barns. The cross section of a single crystal at thermal energies can be very small at room temperature and can be reduced even further by cooling the crystal. Large single crystals are in use both at room and low temperatures as neutron band pass filters[1] to pass thermal neutrons while excluding fast neutrons and gamma rays.

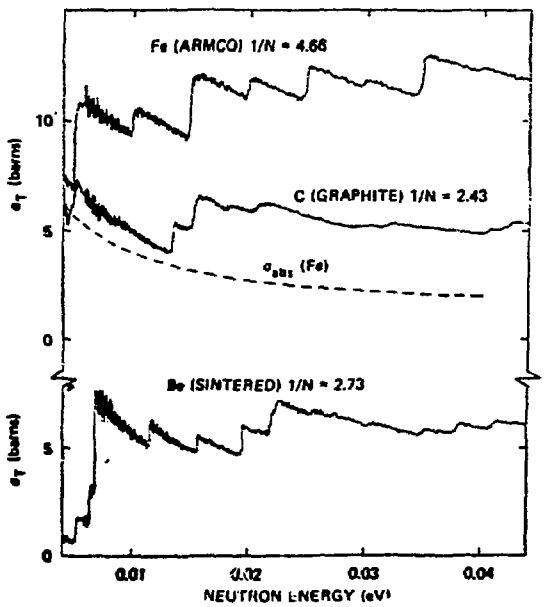

Fig. 1. Total cross sections of polycrystalline Fe, C and Be.

The thermal motion of the atoms in the sample also influences the cross section of a nuclear resonance. In 1937, Bethe and Placzek[2] calculated the Doppler broadening assuming a Maxwellian velocity distribution of gas atoms. In 1939, Lamb[3] calculated the resonance shape for nuclei bound in a Debye crystal. In the weak binding limit where the Doppler width, Δ, plus the natural width, Γ, is >> $2\theta_D$ (θ_D = Debye temperature), the observed resonance shape would be the same as for a gas, but an effective temperature which is somewhat greater than the temperature of the crystal must be used. Lamb also predicted that if an atom were more tightly bound in the lattice, i.e., medium binding where ($\Delta + \Gamma$) ~ $2\theta_D$, some fine structure should appear in the resonance shape. Several experiments have been performed to observe the structure of resonances in both solids[4,5,6] and gases.[6,7] For a crystal at a temperature < θ_D and for a low energy resonance with a small Γ, Lamb showed that a "recoilless" Breit Wigner peak could be expected, as well as structure or general broadening at higher energies. Trammell[8] has pointed out that a detailed study of the 1.057-eV resonance in ^{240}Pu might be able to give detailed information concerning the crystal vibrational spectrum. Recently, Liou and Chrien[9] studied this resonance using both metal and oxide samples.

Experimental Method

The neutron total cross sections of several single crystals (silicon, copper, sapphire), several polycrystalline samples (rolled Cu, armco Fe, sintered Be, graphite, cast Bi and rolled Ta) and a fine powder copper sample were measured from 0.002 to 5 eV. The measurements were made at ORELA using neutrons from the water moderated Ta target with the linac producing ~ 30 nsec pulses at 25 Hz. A 1 cm thick 111 cm dia. ^{6}Li glass scintillator was located at 17.870 m resulting in a neutron energy resolution $\Delta E/E$ = 0.3%. A few measurements were made on the "single" copper crystal with a 25 mm dia. ^{6}Li glass scintillator to reduce the angular divergence of the detected neutrons. The samples were placed at 9 m and the effect of a small rotation of the Cu "single" crystal from the direction of the neutron beam was studied. Samples were alternated with an open beam and background samples every ~ 30 minutes; runs were of 1-2 days duration.

Transmission measurements were also made on ^{240}Pu sample using a 17.858-m flight path at 400 Hz. A Cd filter was used to eliminate overlap neutrons and 2 cm of Pb to reduce the intensity of gamma rays from the target. Measurements were made at room temperature and at 95 K which is much lower than the Debye temperature (~ 175 K). In the 1 eV energy region ~ 40000 counts per channel were collected for a channel width of 0.5 x 10^{-3} eV. The sample was an Al-Pu alloy containing only 0.726% ^{240}Pu with a thickness of ^{240}Pu of 2.12 x 10^{-6} a/b. The uniformity of the sample was ~ 2% over the area used. The Ta metal sample consisted of 15 1.5 mm thick rolled sheets and was measured at 11 K and room temperature using a repetition rate of 25 Hz. The

†Visiting scientist from Nuclear Research Centre-Negev, Beer-Sheva, Israel.

$^{180}Ta_2O_5$ sample was a fine powder and was measured at room temperature and 175 K. Each sample was measured for several days at each temperature.

Data Analysis

The time-of-flight spectra were corrected for the dead-time of the time digitizer, 1.1 μsec, and a constant room background which was ≤1%. Transmissions were computed using the monitor counts from a fission chamber; the total cross sections were computed from the average thicknesses of the samples.

The resonance parameters for the 1.057-eV resonance in ^{240}Pu were obtained by R. F. Carlton using the code SAMMY[10] and by R. R. Spencer using the code SIOB.[11]

Results and Discussion

The total cross sections of polycrystalline Fe, C and Be samples are shown in Figure 1. The scattering cross section for this Armco Fe sample is ~ 9 barns in the thermal energy region and drops to ~ 1 barn at 0.0050 eV, which corresponds to twice the lattice spacing of the 1,1,0 planes for Fe; Fe is body centered cubic with a = 2.86645 A. At lower energies the scattering arises from isotope incoherent scattering (0.4 b) and thermal inelastic scattering; the latter can be reduced by cooling the sample. The other breaks arise from planes with higher Miller indices. Recently, Johnson and Bowman[12] have observed a large number of these breaks at higher energies in samples of rolled Fe.

The scattering cross sections of sintered Be and C (graphite) are ~ 5 b except at low energies. (The absorption cross sections of Be and C are only 10 and 3 mbs.) The breaks in Be at 5.3 and 6.9 meV are well known, but the intermediate one at 6.5 meV has not been reported previously, probably because of insufficient energy resolution. The size of the step depends on the number of atoms in the reflecting plane and the Debye-Waller factor. Above each break the coherent scattering cross section has a 1/E energy dependence. At low energies the thermal inelastic scattering can be reduced to <0.1 b by cooling the sample. Thick filters of polycrystalline materials have been used at reactors to produce beams of "cold" neutrons.

The total cross sections for the copper powder sample and two different measurements on a "single" copper crystal are shown in Figure 2. The scattering is ~ 9 barns for the copper powder but only ~ 2 barns for the "single" crystal. This reduction is consistent with the coherent cross section of 7.5 b. The remainder arises from isotope and spin incoherent scattering (~ 0.5 b), and thermal inelastic scattering. In addition, there are many narrow peaks associated with lattice planes in the single crystal. The widths of the peaks is a measure of the mosiac spread of the crystal, the angular resolution, and the neutron energy resolution. The two lower curves are for the same Cu crystal but oriented differently with respect to the direction of the neutron beam. Additional measurements have been made rotating the crystal by ~ 0.2°, and the peaks shifted appreciably in energy (~ 1 meV) with this small change in angle.

The data for cast Bi are shown in Figure 3. The scattering is ~ 40% of that expected from the free atom scattering of 9.3 barns. The extinction effects are due to the large size of the crystallites in this cast sample. The two low energy peaks at 3.38 and 4.22 meV are only ~ 0.04 meV wide. Since the absorption and the spin incoherent cross sections of Bi are very small, 9 and ~50 mbs respectively, a filter of a cold large single crystal of Bi in a reactor neutron beam would be a valuable technique for eliminating gamma rays and greatly reducing resonance and high energy neutrons from the neutron beam.

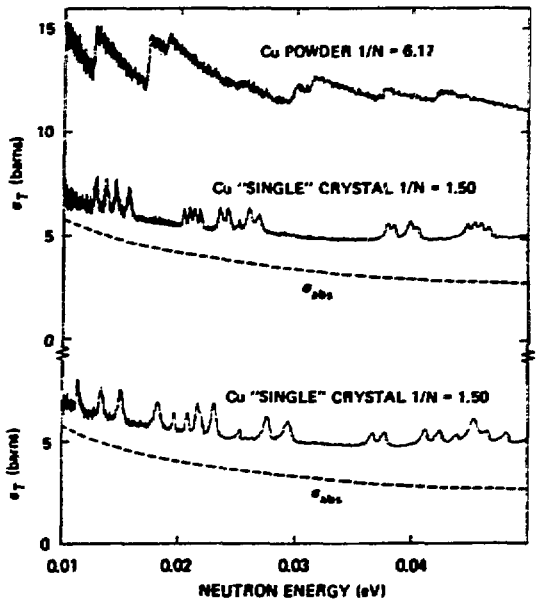

Fig. 2. Total cross sections of Cu powder and two measurements on a "single" Cu crystal.

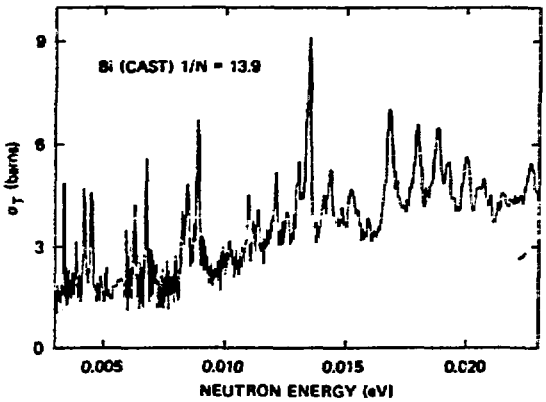

Fig. 3. Total cross section of Cast Bi.

Figure 4 shows the data for single crystals of Si and sapphire. The scattering of Si at thermal energies is only ~ 10% of its value above a few eV energy and arises from incoherent scattering (9 mb) and thermal inelastic scattering. This "perfect" single crystal has a very small mosiac spread; hence, the intensities of any narrow peaks are too small to be observed. The sapphire "single" crystal shows much structure due to the mosiac spread. The gradual increase of the cross section in Figure 5 is due to the Debye-Waller factor and the small broad fluctuations with ~ 50 meV spacing may be due to crystal vibrations.

The transmission of the rolled Ta sample (consisting of 15 1.5 mm thick plates) is shown in Figure 6. The sharp break at 11.3 meV corresponds to scattering from

1,1,0 planes expected for a sample with small crystallites. The sharp peaks are probably due to coherent scattering from lattice planes similar to that observed with the Cu single crystal. The broad structures must be combinations of sharp breaks, overlapping clusters of narrow peaks and possibly phonons.

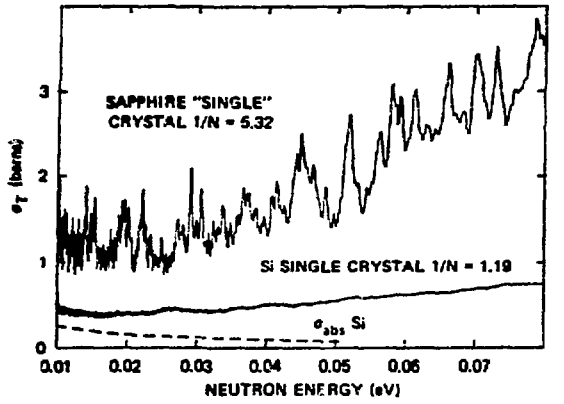

Fig. 4. Total cross sections of Si single crystal and sapphire.

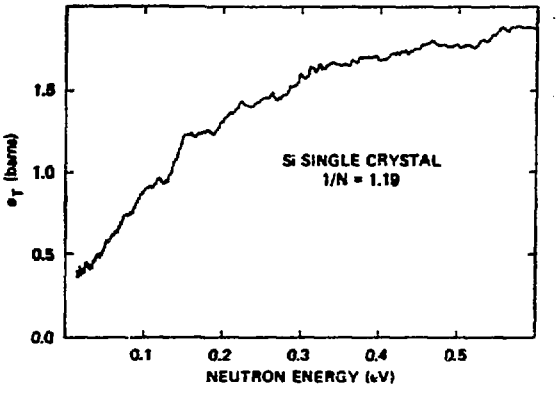

Fig. 5. Total cross section of the Si single crystal.

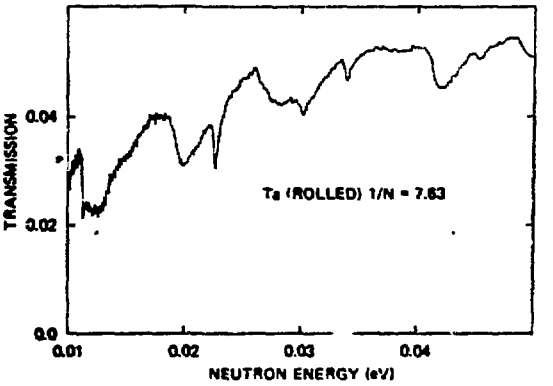

Fig. 6. Total cross section of rolled Ta.

The transmission data for the 1.057-eV resonance in ^{240}Pu are shown in Figure 7 with fits from the code SAMMY using the free gas approximation for the Doppler broadening. The effective temperatures used for the two measurements were 122 K and 300 K based on a Debye temperature of 175 K. The goodness of fit was 0.91 for the cold run and 0.93 for the room temperature run. Both runs gave exactly the same resonance energy of 1.0565 ± 0.0002 eV. Values of 30.6 ± 0.4 meV for Γ_γ and 2.35 ± 0.02 meV for Γ_n were obtained from the cold run and 30.8 ± 0.6 meV and 2.37 ± 0.02 meV from the room temperature run. The uncertainties do not include systematic uncertainties. A 9° K uncertainty of the effective temperature of the cold sample would produce a 0.6 meV uncertainty on Γ_γ. The average value of Γ_γ of 30.7 ± 0.6 meV is somewhat lower than the most recent determination.[3] Including an estimated uncertainty of 2% for the sample thickness, the average value for Γ_n of 2.36 ± 0.05 meV agrees with that contained by Liou.[9]

Trammell[13] has estimated that at 95 K approximately 70% of the absorption by this resonance should be in the undisplaced (Mossbauer) peak and should have the nuclear width Γ. The remaining 30% should be at higher energies and should be Doppler broadened. The recoil energy after neutron absorption by this resonance is 4 meV. At room temperature only 17% would be in the

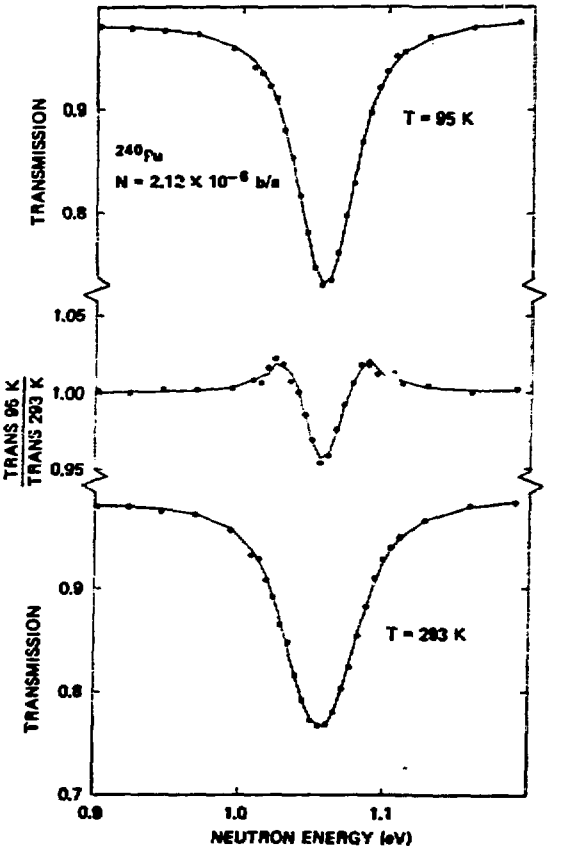

Fig. 7. Transmission of the ^{240}Pu sample at 95 K (top), transmission at 293 K (bottom) and ratio (middle). The statistical uncertainties in the region of the resonance are 0.2% for the transmissions and 0.3% for the ratio.

recoilless Mossbauer peak. The excellent agreement for both temperatures using the gas approximation and the fact that both gave the same resonance energy is not is not consistent with the predicted change in the intensity of the recoilless peak for these 2 temperatures. The middle curve in Figure 7 shows the ratio of the measured transmissions at the two temperatures compared to the ratio of calculations using the gas approximation. Any appreciable change in the intensity in the recoilless peak from the cold to room temperature should produce a noticeable asymmetry. Similar data on the 0.433-eV resonance in ^{180}Ta also did not show any asymmetry.

Conclusions

In the thermal energy region the physical form of a sample can have a large effect on the measured total cross section. Some crystalline materials and "single" crystals can have large self shielding and exhibit narrow peaks arising from the mosaic spread of the crystal. No convincing evidence was found for a recoilless peak for the 1.057-eV resonance in ^{240}Pu or the 0.433-eV resonance in ^{180}Ta.

Research was sponsored by the Division of Nuclear Sciences, U.S. Department of Energy, under Contract No. W-7405-eng-26 with the Union Carbide Corporation.

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