ANOMALOUS NEUTRON DIFFRACTION IN α CADMIUM SULFIDE

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The interaction of neutrons with isolated nuclei may be formulated[1] in a general way to include absorption as well as scattering by expressing the scattering length in complex form. The imaginary component of the complex length or amplitude is directly related to the total cross section and is expected to be small except at neutron energies in the neighborhood of a resonance. The energy dependence of the neutron cross section in the resonance region has been given explicitly in the Breit-Wigner dispersion relations. Although the above formulation of neutron scattering is well accepted, a direct experimental demonstration of the imaginary or 90° out of phase scattering component has not previously been given.

Since the phase of scattered radiation is not directly observable, the effect of the imaginary component can be made apparent only through intensity differences caused by interference. Thus, in the Bragg scattering of neutrons from a noncentrosymmetric crystal, the imaginary component from a resonant nucleus can be detected through interference with a real component which is in phase because it originated from nuclei at appropriately displaced sites. A striking result of such interference is a nonequivalence of diffracted intensity from the opposite sides of a diffracting plane; thus $I_{hkl} \neq I_{\bar{h}\bar{k}\bar{l}}$. This result, which is known as a failure or breakdown of Friedel's law,[2] has been frequently observed in the x-ray case and is directly connected with the anomalous scattering which occurs near an absorption edge. The present investigation clearly establishes the corresponding effect for the neutron scattering case and demonstrates a strong energy dependence of the neutron scattering amplitude.

The first evidence for the anomalous scattering effect with neutrons was obtained with cadmium sulfide, chosen because it contains a strong neutron absorber, the Cd^{113} isotope, and crystallizes with a noncentrosymmetric structure. Thin plates of α-CdS, space group $P6mc$, grown from the vapor phase,[3] were used in the majority of the experiments. The experiments consisted of measurement of integrated intensities of (hkl) and $(\bar{h}\bar{k}\bar{l})$ mates of a series of reflections at several wavelengths. The crystallographic phase relations are such that the (004) and (008) pairs should give equal intensities while the (002), (006), and (00, 10) pairs should give unequal intensities for the case where the cadmium scattering amplitude is complex. Figure 1 gives convincing evidence of Friedel's law failure for the latter group of reflections, strongly implying a complex scattering amplitude and hence anomalous scattering.

Figure 2 gives the ratio of $I_{002}/I_{00\bar{2}}$ plotted against wavelength and illustrates the strong

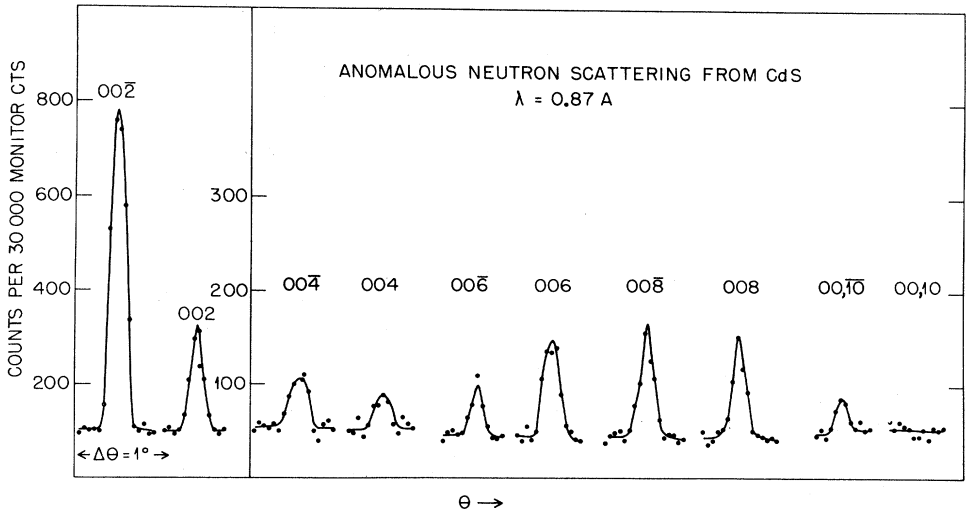


FIG. 1. Neutron diffraction intensities from a single crystal of CdS at $\lambda = 0.87$ A, illustrating nonobservance of Friedel's law in the cadmium resonance region.

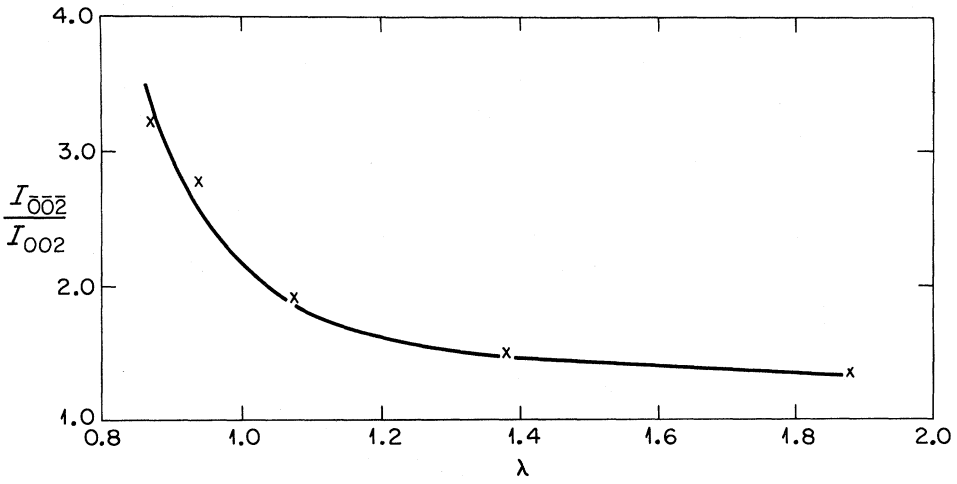


FIG. 2. The wavelength dependence of the intensity ratio $(\bar{0}0\bar{2})/(002)$ of CdS in the cadmium resonance region. The smooth curve is based on the Breit-Wigner formulation. The experimental points are averages of several measurements.

energy dependence of the ratio due to the variation of the complex amplitude of Cd with wavelength. The smooth curve is based on the Breit-Wigner formulation. The coherent scattering amplitude, f, can be expressed in terms of the Breit-Wigner single level parameters in the following way:

$$f = f_0 + \Delta f' + i\Delta f'',$$

where f_0, the nonresonant scattering amplitude, is given approximately by the nuclear radius; the real resonant increment, $\Delta f'$, and the imaginary or 90° phase shifted resonant increment, $\Delta f''$, are

$$\Delta f' = \tfrac{1}{2}\frac{gw\lambda\!\!\!^{-}_0\Gamma_n(E-E_0)}{(E-E_0)^2+\frac{1}{4}\Gamma^2}, \quad \Delta f'' = -\tfrac{1}{4}\frac{gw\lambda\!\!\!^{-}_0\Gamma_n\Gamma}{(E-E_0)^2+\frac{1}{4}\Gamma^2}.$$

Here g is the spin weighting factor, w the isotopic abundance factor, $\lambda\!\!\!^{-}_0$ the wavelength at resonance divided by 2π, Γ_n the neutron width at resonance, Γ the total width, E the energy of measurement, and E_0 the resonance energy.

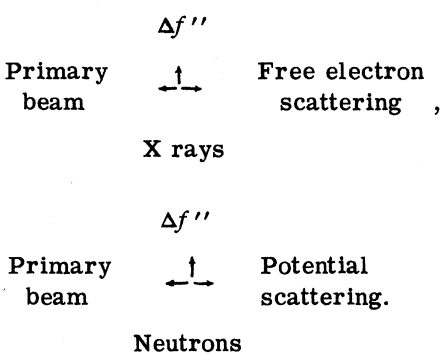
Table I compares experimental values of $\Delta f'$ and $\Delta f''$ with the calculated ones. The resonance parameters Γ, Γ_n, E_0, and g were taken from the cross-section measurements of Brockhouse.[4] It should be noted that the abundance of Cd^{113} in normal Cd is only 12.3%, which correspondingly reduces the magnitude of the resonance terms in the scattering amplitude.

In order to establish the direction of the phase shift, comparisons were made of the intensity inequalities with both x rays and neutrons for the same crystal of CdS. The result, that the direction of the phase shift is the same for x rays and neutrons (indicated in the diagram below), is in agreement with expectation.

Table I. Cadmium scattering amplitudes in units of 10^{-12} cm.

	$\Delta f''$		$\Delta f'$	$f_0+\Delta f'$	
λ in A	Calc	Obs	Calc	Calc	Obs[a]
0.87	0.230	0.21	-0.280	0.34	~0.35
0.94	0.178	0.16	-0.265	0.36	~0.35
1.075	0.125	0.12	-0.238	0.38	~0.40
1.38	0.085	0.08	-0.206	0.41	~0.40
1.88	0.067	0.06	-0.184	0.44	~0.40

[a]A value of $f'(\mathrm{Cd}) = 0.38$ at $\lambda = 1.13$ has been determined previously from powder data of CdO by B. N. Brockhouse and A. T. Stewart according to a private communication.



Anomalous x-ray scattering has had important applications in crystallographic work to the determination of absolute configuration[5] and to the direct solution of noncentrosymmetric crystal structures.[6,7] Similar applications of anomalous

neutron scattering are clearly indicated. The potentially useful anomalous scatterers include Li^6, B^{10}, Cd, and a number of examples in the rare earth and actinide series. Experiments are under way to investigate Li^6 and B^{10} as well as several others.

*Operated for the U. S. Atomic Energy Commission by Union Carbide Corporation.

[1]See for example O. R. Frisch, Progress in Nuclear Physics (Academic Press, Inc., New York, 1950), Vol. 1, Chap. 8.

[2]R. W. James, The Optical Principles of the Diffraction of X Rays (G. Bell and Sons Ltd., London, 1954), Chap. II, p. 33.

[3]The authors are indebted to D. R. Boyd and Y. T. Sihvonen of the General Motors Research Laboratories for the CdS platelets.

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THEORY OF THE PULSATION OF FLUORESCENT LIGHT FROM RUBY

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It has been observed[1,2] that when one pumps a ruby crystal placed between two parallel reflecting end plates with intense light in order to create an excess of population in the fluorescent R level(s), there is a threshold of pumping power above which (a) the light from the R line is emitted in pulses accompanied by (b) a sudden spatial and spectral narrowing. This pulsed character of the output is not accounted for by the theory of Schawlow and Townes[3] for a gas Fabry-Perot type maser. We propose here a different approach to this induced fluorescence which predicts that as long as the pump power is above a certain threshold, part of the fluorescent power will occur in recurrent bursts or pulses. From the theory we derive quantitative estimates of the pulse repetition rate, the fraction of power in the pulses, and the nature of the output between pulses, all in terms of the pump power and the ordinary properties of the crystal and end plates. The equations connecting the spectral and spatial narrowing with the time-varying behavior will be developed.

We consider two infinite parallel plates of power reflection coefficient R and a distance d apart. The space between is filled with a ruby crystal. The crystal will be assumed just inhomogeneous enough and the end plates rough enough so that after many internal reflections photons will not interfere in any regular manner; therefore, we may use a straight photon model of the light. Diffraction effects will be assessed in a later article. We shall employ an optically thin crystal which can be pumped uniformly and in which the fluorescent photon density does not vary appreciably across the crystal, i.e., $1-R \ll 1$. To describe the radiation then, we may use the density of photons $u(\nu, \theta)$ traveling in the crystal at a frequency ν from the line-center frequency (measured in units of the half-width at half power) and at an angle θ to the norm. (All photon and population numbers will be measured per unit area of end plate.) We shall assume that we are dealing with a single component of the fluorescent R lines with an approximately $\cos^2\theta$ radiation pattern around a crystal axis that is normal to the end plates. (In an actual apparatus the crystal alignment need not be perfect if the crystal sides are smooth and parallel enough to form a light-pipe between the end plates.) The gain (negative absorption) coefficient $\alpha(\nu, \theta)$ associated with the R line will be assumed to result from a homogeneously broadened line and therefore may be written $[(1+\nu^2)N]^{-1}(n_2 - n_1)A\cos^2\theta$ where the peak absorption coefficient A for ruby is commonly of the order 0.4 cm^{-1} at room temperature and 10 cm^{-1} at liquid-nitrogen temperature. We shall measure the level populations and the difference $n = n_2 - n_1$ in units of the difference n_0 required for the maser material gain at $\nu, \theta = 0$ to just balance the end-plate losses. $n_0 = N(1-R)/Ad$. N is the total number of ions (per unit area) which when measured in units of n_0 will be called M. We shall also measure, for convenience, the