PHYSICAL REVIEW VOLUME 82, NUMBER 6 JUNE 15, 1951

Letters to the Editor

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The Index of Refraction for Neutrons*

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(Received May 1, 1951)

IN a recent paper on variational principles in scattering theory, Lippmann and Schwinger[1] have treated the scattering of slow neutrons by bound protons as an application of their time independent formulation, which they obtain from time dependent scattering theory by means of several devices including the adiabatic reduction of the interaction to zero at $t=\pm\infty$. The merits of this formulation are (a) the exact transition rate is obtained in the form $\omega_{ba}=2\pi\delta(E_b-E_a)\,|T_{ba}|^2$, where T is an operator which in Born approximation is the interaction energy; (b) a variational principle is given for the T-matrix; (c) a sum rule is given which states that the total rate of transition out of the initial state Φ_a is $-2\,\mathrm{Im}T_{aa}$.

By a simple generalization from the neutron scattering by one bound proton we have considered the scattering by a thin slab of material containing many nuclei. The Φ_a component of the final state $S\Phi_a$, where S is the Heisenberg S-matrix, is $[1-i(m\lambda/2\pi)T_{aa}]\Phi_a$. When the sum rule is invoked, the imaginary part of T_{aa} gives the customary amplitude extinction factor $(1-\frac{1}{2}N\sigma x)\approx\exp(-\frac{1}{2}N\sigma x)$, where x is the thickness of the slab, and N is the nuclear density. Similarly, the real part of T_{aa} represents a phase shift, which one might interpret as due to the index of refraction of the slab. When the Fermi approximation to T_{aa} is used, the phase shift is in agreement with the usual formula for the index of refraction, $n=1-N\lambda^2a_c/2\pi$, where a_c is the isotopic average of the *bound* coherent scattering amplitudes of the nuclei.

One cannot expect an actual derivation of the index of refraction to follow from time dependent scattering theory, because from the time dependent viewpoint $S\Phi_a$ is the final state after the interaction has been adiabatically reduced to zero, and it is not clear why the phase shift in $S\Phi_a$ should necessarily be the same as that which the interaction would produce in a stationary wave function. To overcome this objection we have found a formal solution to the stationary eigenvalue problem identical with the $\Psi_a^{(+)}$ of Lippmann and Schwinger, which we show represents an incoming plane wave and an outgoing scattered wave. This stationary formulation is similar to that of Moller,[2] who introduced the S-matrix into stationary scattering theory. The connection between the S-matrix and the wave function is that the outgoing part of $(S-1)\Phi_a$ is identical with the scattered wave. On the basis of this theory we arrive at a general derivation of the index of refraction for gases, liquids, and solids.

This derivation of the index clearly shows that there is no doppler effect due to the motion of the individual nuclei, because the λ in the formula is the neutron wavelength relative to the boundary of the slab. The variational principle gives, besides the Fermi approximation, a temperature dependent correction to the simple formula, thereby taking into account the effect of virtual inelastic interactions of the neutron with the material. We have not as yet evaluated the correction in a specific case, but Lippmann[3] finds a correction of 0.2–0.3 percent to the scattering cross section of parahydrogen in its lowest rotational state for neutrons of zero energy. In liquids or solids at fairly high temperature and for neutrons of nonzero energy it would seem possible for the correction to be of the order of 1 percent. A change in the critical angle of reflection of a mirror of this amount would be significant in the experiment of Hughes, Burgy, and Ringo,[4] and might cause a detectable change in the intensity of a reflected beam of pile neutrons.

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* Research carried out under contract with the AEC.
[1] B. A. Lippmann and J. Schwinger, Phys. Rev. 79, 469 (1950).
[2] C. Moller, D. Kgl. Danske Vidensk. Selskab, Mat-fys. Medd. XXIII, Nr. 1 (1945).
[3] B. A. Lippmann, Phys. Rev. 79, 481 (1950).
[4] Hughes, Burgy, and Ringo, Phys. Rev. 77, 291 (1950).

On the Ferroelectric Curie Point of Tungsten Oxide

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(Received April 9, 1951)

SINCE Nagasawa[1] pointed out the possible existence of ferroelectric phenomena in tungsten oxide, the physical properties of this substance have been studied extensively at several laboratories in Japan. Matthias[2] also reported independently the experimental investigations of the ferroelectric properties of tungsten oxide. According to the preceding investigators, this substance was found to have many of the properties peculiar to ferroelectrics, i.e., the very high value of permittivity, the characteristic variation with a biasing field, the hysteresis loop, the domain structure, and the sensitive variation with external stress. On the other hand, the domain structure has not been influenced by an electric field, and its transition temperature has not been discovered up to about 550°C, which state of affairs makes us hesitate to conclude that tungsten oxide has ferroelectric properties. In order to clarify this situation, we have recently extended our experimental observations to higher temperatures and have found that tungsten oxide really has a spontaneous polarization with the Curie point at about 710°C. In the following the results of our observations are reported briefly.

The domain structure of a single crystal of area 0.01 mm^2 and thickness 0.01 mm was observed, up to 950°C, by a polarized-light microscope, using a very small electric furnace. On heating the crystal, the domain structure did not vary at all up to about 710°C, at which temperature the domain patterns vanished with the field of view becoming dark almost suddenly, and at still higher temperatures the birefringence was no longer discernible. On cooling, the domain patterns appeared almost suddenly at about 685°C, and the original features were approximately realized again at room temperature.

Next we prepared a rectangular crystal (0.4×0.2×0.02 mm) with a few bands inclined by 45° to the edges, which had two fired-on silver paste electrodes separated by 0.1 mm along the longest direction of the crystal. It was then connected to the 100-v terminals of a battery through a 20-kΩ rheostat, the resistance of the crystal being 2.5 kΩ. On account of the rather high conductivity, the temperature of the crystal is raised by the Joule's heat by decreasing the resistance of the rheostat, and at about 550°C the new 45° bands begin to appear. As the temperature is raised more new bands appear, and finally at about 700°C the whole crystal becomes dark suddenly. The successive disappearance of the 45° bands is observed as the temperature is lowered, i.e., as the resistance of the rheostat is increased, although nearly half of the newly-appeared 45° bands still remain at room temperature. It is shown by x-ray analysis that the a- and b-axes of a plate crystal lie parallel or perpendicular to the edges in the plane of the plate, and hence the abovementioned results can be clearly understood by assuming that tungsten oxide has a spontaneous polarization with Curie point at 710°C in the direction of the b-axis, and further by taking into account the generally accepted fact that the coersive field decreases with increasing temperature. The results of the x-ray analysis of the crystal structure suggest to us that the spontaneous polarization does not lie along the direction of the *a*-axis, while that along the *c*-axis may perhaps exist. We have been unable to ascertain clearly the *c*-axis polarization, since any plate crystal with the *c*-axis lying in the plane of the plate can, unfortunately, not be prepared.

By using a differential dilatometer, we made relative measurements of the thermal expansion of tungsten oxide with respect to that of barium titanate, both samples being ceramic rods, and found that a contraction of volume of tungsten oxide was clearly observed at about 710°C on heating, and at about 685°C on cooling, respectively. Furthermore, by cooling ceramic powder of tungsten oxide, an anomaly was distinctly observed near 700°C on the cooling curve, which fact suggests to us the existence of a latent heat or a λ-type anomaly of the specific heat at this temperature.

The low resistance of tungsten oxide makes the dielectric measurements quite difficult, especially at high temperatures. Although any decisive result, therefore, has not yet been obtained from our measurements of both the permittivity and the hysteresis loop, an anomaly of the permittivity in the neighborhood of 700°C and the almost linear variation of polarization with applied field above this temperature have been qualitatively observed to exist.

From these observations we may conclude that the tungsten oxide actually has ferroelectric properties and that its Curie point is situated at about 710°C. Quantitative observations are now in progress, the results of which will be reported shortly.

The authors wish to thank Professor T. Muto for his kind criticism and advice.

[1] S. Nagasawa, Denki-kagaku **16**, 13, 57 (1948); **17**, 174 (1949).
[2] B. T. Matthias, Phys. Rev. **76**, 430 (1949).

Gamma-Rays Emitted in the Disintegration of the 36-Hour Rh^{105}

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(Received May 2, 1951)

THE radiations of the 36-hour Rh^{105} have been examined by several groups of investigators,[1] and more recently by the writers.[2] Subsequent to all of the abovementioned measurements, more precise data have been obtained by Duffield and Langer.[3] They did not, however, find any evidence of the gamma-ray at ~0.3 Mev which had been observed by Sullivan, Sleight, and Gladrow[1] and by the writers.[2] Accordingly, a second quantity of metallic ruthenium has been irradiated by slow neutrons for ten hours in the Oak Ridge pile. The 36-hour rhodium was thus grown from its 4.5-hour ruthenium parent element.

The ruthenium used for pile irradiation was purified before exposure by distillation of RuO_4 from a perchloric-sulfuric acid solution of chemically pure $RuCl_3$. Irradiated Ru was dissolved in a mixture of 10 parts conc. HCl, 1 part conc. HNO_3, at 160°C. Rhodium carrier was added to the Ru solution and RuO_4 was distilled from a perchloric-sulfuric acid solution. Inactive $RuCl_3$ was added to the residue and another RuO_4 distillation effected. The residue in the distillation flask was diluted to 1 *M* in acid and treated while hot with magnesium powder to precipitate rhodium. After dissolving excess magnesium in HCl, the suspension was filtered, the rhodium washed with HCl, hot water, alcohol, and ether, then dried.

The purified source of Rh^{105} was placed between two blocks of aluminum, each having a surface density of 1.7 g/cm². This thickness was more than sufficient to stop all of the beta-rays of Rh^{105}, because they have a maximum energy of only 0.570±0.005 Mev.[3] Adjacent to either aluminum block was placed a G-M tube, having glass walls of thickness 0.1 cm and copper cathodes of wall thickness 0.013 cm. In one counter, the half-period of the quantum radiations being emitted by the source of Rh^{105} was measured; with the other, absorption curves in lead of the gamma-rays were obtained. The decay curve of the gamma-ray activity of Rh^{105} was followed for three half-periods, and its slope gave a half-period of 35±1 hours. Two lead absorption curves for the gamma-rays of Rh^{105} are shown in Fig. 1. The first of these curves was taken at the beginning of the decay curve, and the second curve was obtained forty hours later. The quantum energy taken from the slope of the curves is 0.3 Mev.

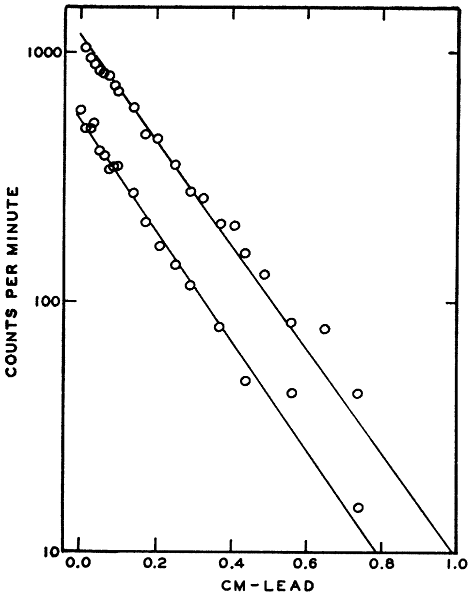

FIG. 1. Absorption in lead of the quantum radiations emitted in the disintegration of rhodium (105). The two curves, separated in time by forty hours, indicate a quantum energy of 0.3 Mev.

The question arose whether the presence of quantum radiations might be accounted for by bremsstrahlung effects. This seemed to be an unlikely explanation for the presence of the gamma-rays, because the mean energy of the beta-rays from Rh^{105} is only 0.2 Mev.[3] However, to show positively that the results are not in any way related to the presence of bremsstrahlung, comparative measurements were carried out, using the beta-spectrum of RaE. The source of Rh^{105}, the gamma-ray activity of which had been followed for three half-periods, was placed before a thin window "bubble" counter. The beta-ray counting rate was observed, and after 0.1 cm of lead had been placed before the counter, the gamma-ray activity was likewise noted. The source of Rh^{105} was removed; a source of $Ra(D+E+F)$ was positioned to give the same beta-ray counting rate as was previously observed for Rh^{105}, and the same sheet of lead was placed over the counter to observe the residual counting rate. The data are given in Table I, where it is clear that although the counting rate from $Ra(D+E+F)$ did not exceed the background count, that of Rh^{105} was three times greater. Were the quantum radiations which were detected in the counter originating from bremsstrahlung production in the lead sheet, the counting rate from $Ra(D+E+F)$ should have been the larger, because the beta-rays of RaE have a maximum energy of 1.17 Mev.

TABLE I. Gross counting rates (per minute).

Background	$Ra(D+E+F)$	Rh^{105}
37.5±3	39±4.5	116±4

As previously reported,[2] beta-gamma coincidences have actually been observed in the disintegration of Rh^{105}. In conclusion, it can be stated that data taken with two different sources of Rh^{105}, produced at different times and from different samples of target material, indicate that gamma-rays are present in the disintegra-