# EVIDENCE FOR A COOPERATIVE ELECTRON TRANSITION IN PLUTONIUM AT LOW TEMPERATURES

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The temperature dependence of the elastic moduli and ultrasonic attenuations of plutonium is not consistent with the antiferromagnetism hypothesis for this metal at low temperatures. The elastic and anelastic behavior of plutonium indicates the possible existence of a cooperative electron transition at 66°K.

Numerous investigations, many of which were reported at two conferences on plutonium,[1,2] have been devoted to the study of the behavior of this metal at low temperatures. The alpha (monoclinic), beta (body-centered monoclinic), and delta (fcc) phases of plutonium were found to exhibit a maximum in their temperature dependence of the electrical resistivity in the vicinity of 100°K[1-3] followed by a negative coefficient of resistivity with increasing temperature. The most prominent among the proposed mechanisms to explain this behavior were (1) an antiferromagnetic transition,[4,5] and (2) a phonon-assisted interband scattering of the electrons.[6]

Indirect experimental evidence for antiferromagnetism in plutonium was implied from the results of radiation damage measurements and Hall effect,[2] thermoelectric power,[7] and transverse magnetoresistivity.[8] But neutron diffraction,[9] magnetic susceptibility,[1] and specific-heat measurements[2] failed to support this hypothesis.

The Landau et al. phenomenological theories of second-order phase changes[10,11] require the existence of anomalies in the elastic and anelastic properties in the vicinity of a magnetic transition.[12,13] The objective of the present study was to search for anomalies in these properties in plutonium by employing a sensitive ultrasonic technique.

The simultaneous determination of the longitudinal and transverse sound-wave velocities and their respective attenuations was carried out by means of an ultrasonic pulse technique at a frequency of 10 MHz. Experimental details and the method of data processing will be described elsewhere.[14] The estimated experimental error in the determination of the elastic moduli is 0.2%. The relative ultrasonic attenuations were determined to within better than 1%. The plutonium metal loaned through the courtesy of the French Atomic Energy Commission was electrorefined by the fused-salt method.[1] From a purified cylinder (about 300-ppm impurities by weight) two flat and parallel disks, 12 mm in diameter by 5 mm thick, were cut and hand-lapped to a parallelism of faces to within $2\times10^{-3}$ mm. The room-temperature density was found to be 19.62 g cm$^{-3}$. The temperature variation of the acoustic path length was corrected by using an average linear coefficient of thermal expansion[15] of 55 $\times10^{-6}$ deg$^{-1}$. Ultrasonic measurements were performed in the temperature range between 4.2 and 300°K. Scatter between the sets of data obtained from both disks was within the limits of the experimental error.

The electrical resistivity of plutonium increases with holding time (several thousand hours) at temperatures near 4.2°K. This process is due to self-irradiation damage.[2] It was of interest to

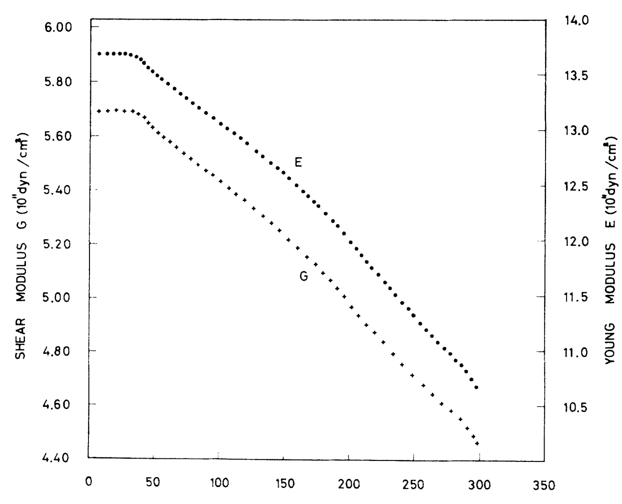


FIG. 1. The temperature variation of the Young ($E$) and shear ($G$) moduli of plutonium.

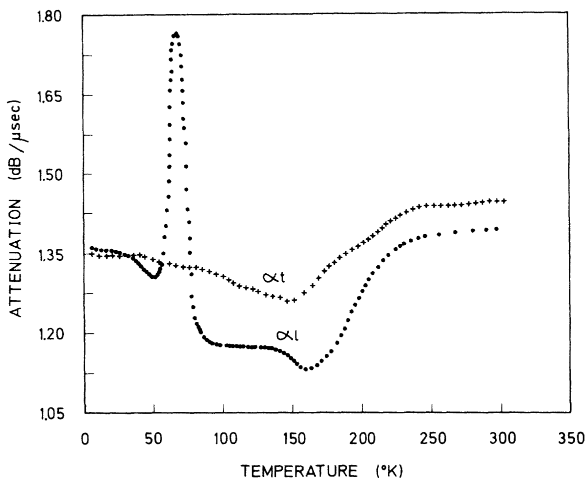


FIG. 2. The temperature variation of the longitudinal ($\alpha_l$) and transverse ($\alpha_t$) attenuation of plutonium.

find out whether the self-damage process affects the elastic and anelastic properties of plutonium under the experimental conditions of the present study. One specimen was held for about 6 h (compared with a 30-min stay in a regular run) before performing the ultrasonic measurements during warming up to room temperature. Neither the elastic moduli nor attenuations were affected.

The Young ($E$) and shear ($G$) moduli of plutonium, Fig. 1, approach 0°K with zero slope, in agreement with the third law of thermodynamics. The temperature variation of the elastic moduli is in accord with the measurements of Taylor et al. (p. 162 of Ref. 2), rather than with those of Lallemont and Solente (p. 147 of Ref. 2). The Debye temperature, calculated from the measured acoustic velocities, and extrapolated to absolute zero, is 206°K. The absence of any anomalies in the temperature dependence of the elastic moduli in plutonium (Fig. 1) indicates that the criteria of Landau-Lifshitz theory[10] are not satisfied. Thus, the antiferromagnetic model for plutonium does not seem to be adequate.

The most significant result of the present study is the appearance of a sharp anomaly, peaked at 66°K, in the longitudinal wave attenuation ($\alpha_l$), Fig. 2. The transverse attenuation ($\alpha_t$) in this temperature range, between 4.2 and 150°K, varies quite smoothly. Pippard[16] has shown that an increase in the longitudinal attenuation only can be due to changes in the electron band population. The behavior of $\alpha_l$ in plutonium suggests, therefore, a cooperative electron transition. The peak of $\alpha_l$, at 66°K, indicates the point where the band overlap is maximal. The increase in both $\alpha_l$ and $\alpha_t$ at temperatures above 150°K is apparently due to a phonon-assisted mechanism.[6]

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