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SOME CONSIDERATIONS ON THE INFRARED ABSORPTION SPECTRUM OF PuO_2 POWDERS AND SINTERED PELLETS

by

L. MANES, A. BARISICH and K. BUIJS

1968

Joint Nuclear Research Center
Karlsruhe Establishment - Germany

European Institute for Transuranium Elements

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SUMMARY

Infrared absorption measurements have been performed on PuO_2 powders and sintered pellets in the spectral range of 4 to 50 μ. A resonance frequency has been found at 360 ± 10 cm^{-1}, attributable to one-phonon fundamental absortion ("Reststrahlen" absorption). On the short wave length side of this absorption band a detailed structure is found, attributable to many-phonon combinations. A remarkable similarity is found between the spectrum of PuO_2 and those of UO_2 and ThO_2. A discussion is given of the effect of sizes and shapes of the crystals on the one-phonon resonance frequency, in the light of recent theories.

KEYWORDS

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SINTERED MATERIALS
FREQUENCY
PHONONS
RESONANCE
LATTICES
GRAIN SIZE

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INTRODUCTION

Our Institute is engaged in a research program on the properties of transuranium elements and their compounds, both from a technological and a fundamental point of view. PuO_2, a compound which is of engineering interest for the development of fast reactor fuels, is the object of rather intensive studies. From a fundamental point of view, its thermodynamical, structural, and transport properties are being investigated.

The present report deals with the study of the optical properties of PuO_2 in the infrared region. Apart from the obvious technological interests in knowing the dispersion properties of PuO_2, one may take full advantage of the rather recent developments of the theory concerning the lattice dynamics of the simplest lattice structure to obtain a good understanding of the vibrational characteristics of this oxide.

The most frequently employed methods to study optical properties of rather opaque solids are either reflection studies on single crystals, or reflection and absorption measurements on deposited thin films of the material.

Unfortunately, notwithstanding considerable research on this line, no PuO_2 single crystals of adequate size have so far been prepared. Reflection measurements on powders or polycrystalline thin specimens suffer from the fact that the real state of the surface may not be controlled adequately, especially in materials as PuO_2 which present a wide range of stoichiometry. A similar type of criticism applies to the use of deposited layers.

We have felt therefore that direct absorption measurements on powders and on sintered pellets could bring more meaningful information. In addition, measurements performed on the same system in both forms provide a useful check. Powders may be investigated by the widely employed method of dispersing them in adequately transparent matrices.

Manuscript received on November 11, 1967.

A fundamental objection(1) has been made recently, however, against optical measurements performed with polycrystalline materials in which the size of the crystals can be very small compared to infrared wave lengths. In interpreting the data, it is necessary in this case to account for the sizes and shapes of the crystallites. To have an idea of the type of information that can be derived from this kind of measurements, we have studied also powders and sintered pellets of UO_2. Since for this material very accurate single-crystal data are available, especially from the work of J.D. AXE and G.D. PETITT (1), a useful comparison may be established. A short discussion on this subject will be presented.

EXPERIMENTAL

Powders of PuO_2 have been prepared by precipitation of plutonium oxalate from acid solutions of oxalic acid, and calcination of the oxalate at 900°C. The O/M ratio was checked by gravimetric methods. The samples used for the optical experiments were always stoichiometric. Spectrographic analysis on the powders showed very low contents of metallic impurities, and a carbon content always less than 50 ppm.

A second method of powder preparation was also employed. A highly pure National Bureau of Standards Pu metal rod was slowly oxidized under oxygen and its O/M ratio was determined. We found no difference in the optical properties between the differently prepared powders, which fact gives reasonable evidence that no detail of the spectrum was attributable to spurious effects related to the method of preparation.

All the preparative and analytical work was performed by the laboratories of ceramics and analytical chemistry of the Institute.

Powdered samples were mostly examined in CsBr dispersion. CsBr-pellets were prepared by weighing out different concentrations (ranging from 0,3 % to 5 %) of PuO_2 in MERCK spectroscopic grade CsBr. The mixtures of PuO_2 and CsBr were ground in a standard steel mill with standard steel balls. A micrographic inspection of the resulting powders, after dissolution of CsBr in water, showed that the greater part of the particles had a diameter of less than 10 μ.

The mixtures were thoroughly dried under vacuum at 300°C for periods of 20 to 40 hours, and then pressed in a Perkin-Elmer standard press form for 13 mm diameter pellets. CsBr was found to require a rather high pressure (13 to 15 tons/cm^2) to give transparent and homogeneous pellets.

Sintered pellets were prepared by our ceramics laboratory using a standard procedure. Sintering occurred under slightly reducing

atmosphere at about 1600°C for three hours. The resulting pellets, measuring about 10 mm in diameter and 1 mm in thickness, were then submitted to a heat treatment to condition their stoichiometry. O/M determinations and purity analyses were duly performed.

Grinding of these pellets to adequate thickness presented a rather difficult problem. Due to the different heat treatments, very often internal cracks hampered the procedure. Besides, due to the necessity of performing the work in glove boxes and under nitrogen, manual procedures of grinding which are common in metallurgical work, had to be discarded. Instead, a Lapmaster 14 machine was employed. The abrasive consisted of a rather fine Al_2O_3 powder (~20 µ grain size). Samples were glued on a perfectly plane metallic support and slowly ground down. The grinding was followed by means of a micrometer. Thicknesses from 40 to 100 µ were thus obtained.

After removal from the metallic support and thorough washing, the thicknesses of the supposedly cylindrical samples were determined by weighing. A precision of about 10 % in the thickness may be claimed. The samples were then glued to metallic frames, to be examined. UO_2 powders and sintered pellets were prepared by techniques similar to those described above.

Due to the high toxicity of plutonium, all operations had to be carried out in glove boxes which considerably hampered a number of manipulations.

A Perkin-Elmer 225 far infrared spectrophotometer, ranging from 4000 to 200 cm^{-1}, was used to perform the absorption measurements. Spectra were run at room temperature.

In order to position the samples in the light beams without any contact with the external atmosphere, a cell was built which was permanently connected to the glove box (fig. 1). 2 mm thick, optically worked, plan-parallel CsBr windows were used up to about 500 cm^{-1}. For the longer wave lengths, to avoid considerable absorption by the windows, a different technique was employed. The samples were sealed in polyethylene envelopes, shown in fig. 2.

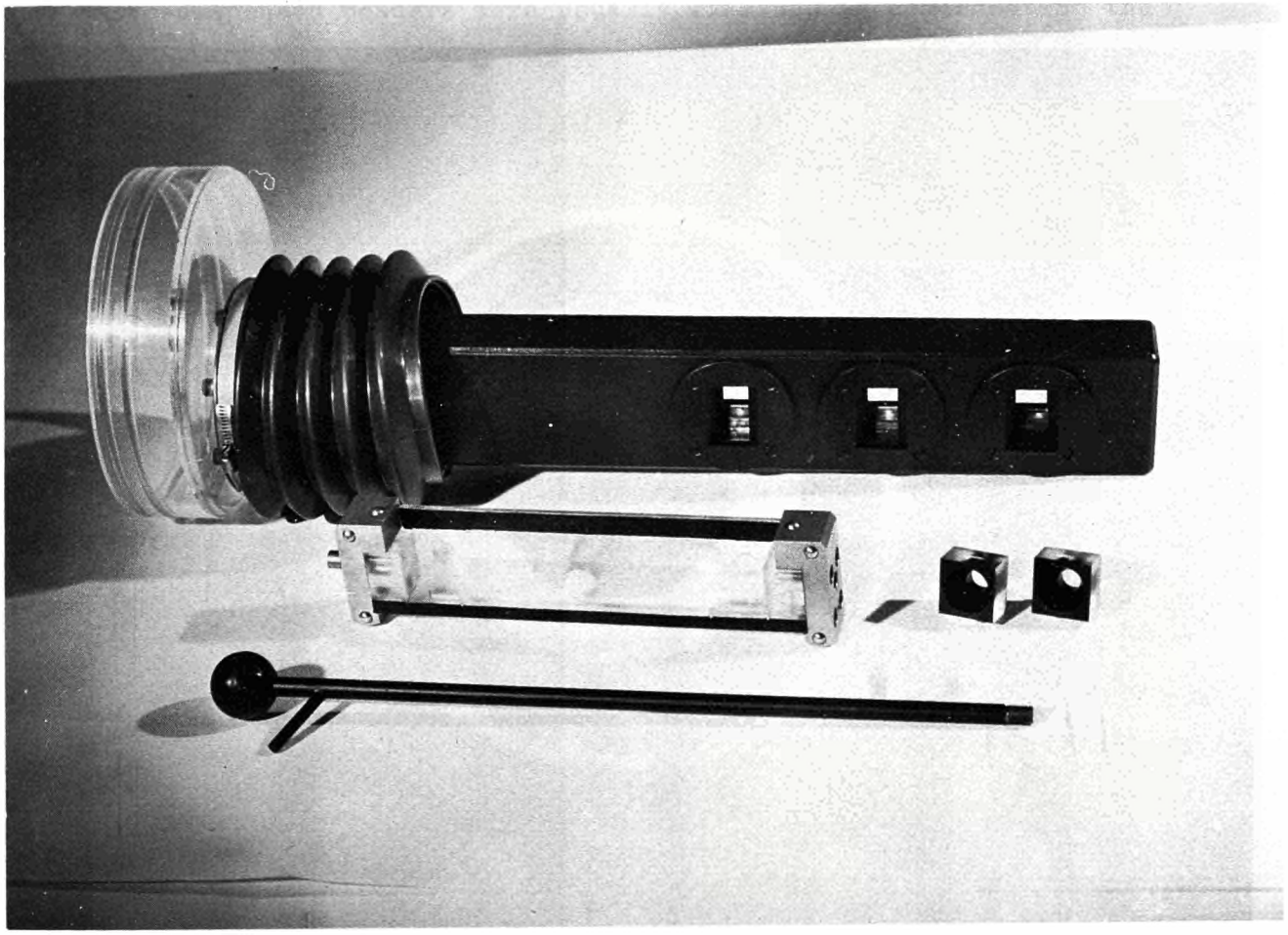

Fig. 1 Cell and sample holder

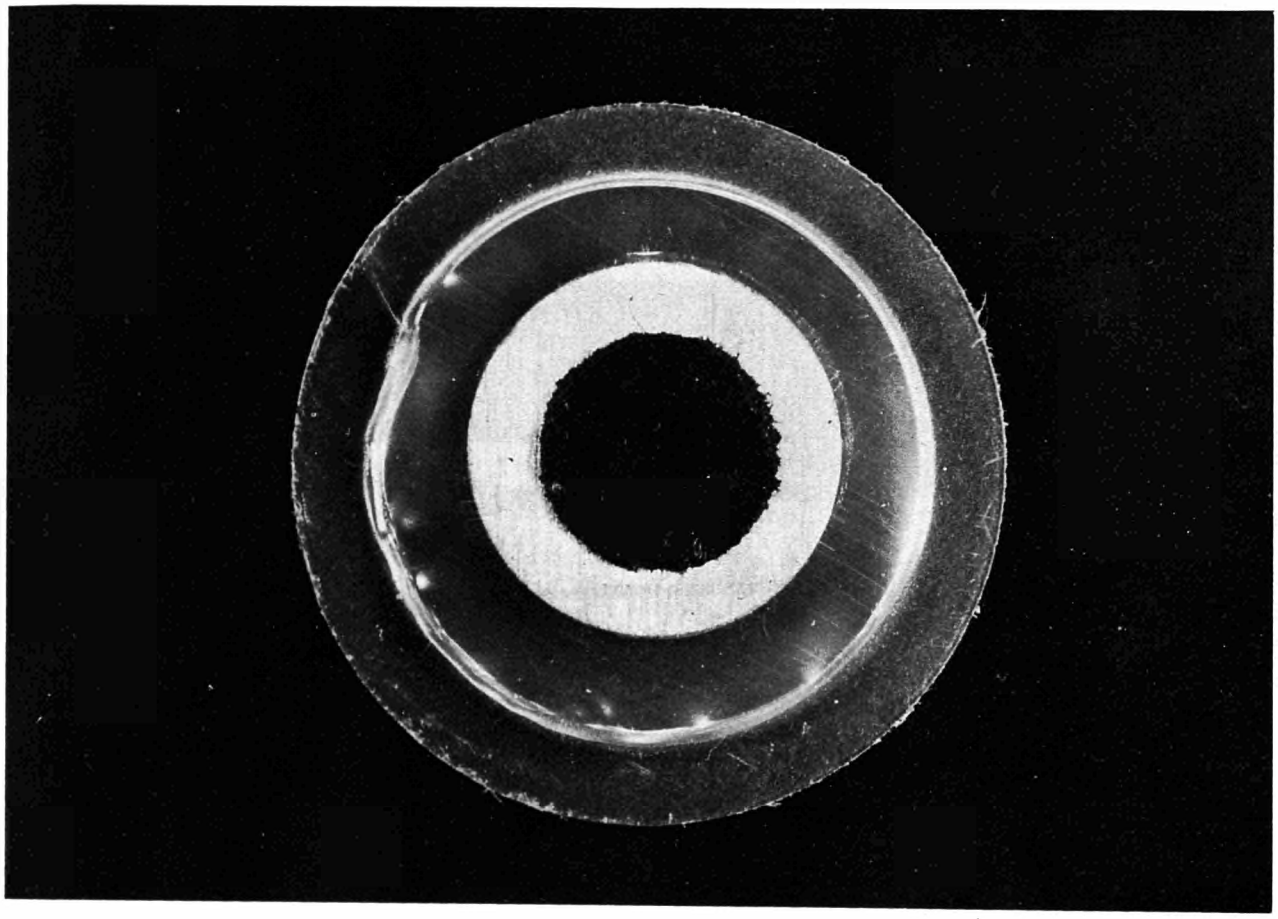

Fig. 2 Polyethylene envelope containing a PuO_2 sintered pellet

With much care, these envelopes could be positioned in the optical beams.

A certain number of interference maxima resulting from the thin polyethylene foils were found to be superimposed on the spectrum. It was very easy, however, to correct for them.

CsBr-dispersed powders were usually examined by compensating the CsBr absorption with a CsBr pellet of comparable thickness in the reference beam; CsBr purity was repeatedly checked.

RESULTS

Fig. 3 shows two typical spectra of PuO_2 and UO_2 dispersed in a CsBr matrix. The concentration is about 0,3 % in weight. The value of the absorption maximum is indicated in the spectrum.

Fig. 4 shows the results on PuO_2 sintered pellets, in the short wave length part of the absorption maximum. The UO_2 spectrum is very well in agreement with the results of BATES (2) and of J.D. AXE (1) on a single crystal. At frequencies lower than 500 cm^{-1} it was difficult to measure accurately due to the strong absorption by the sample.

As shown in the figure, PuO_2 presents a spectrum in many respects very similar to UO_2, although a rather more detailed structure can be found. In fig. 4 the spectrum of a very concentrated CsBr pellet (5 %) is shown. The typical peak at 1030 cm^{-1} is clearly evident, in good agreement with the polycrystalline samples.

Figures 5 and 6 show the values of absorption coefficients both for UO_2 and PuO_2, not corrected for reflection losses. Since a large amount of scattering is to be expected, these values for UO_2 are higher than those measured on single crystals. They are, however, in good agreement with the results of H.P. MYERS and J.A. GYLLÅNDER (3) on sintered pellets.

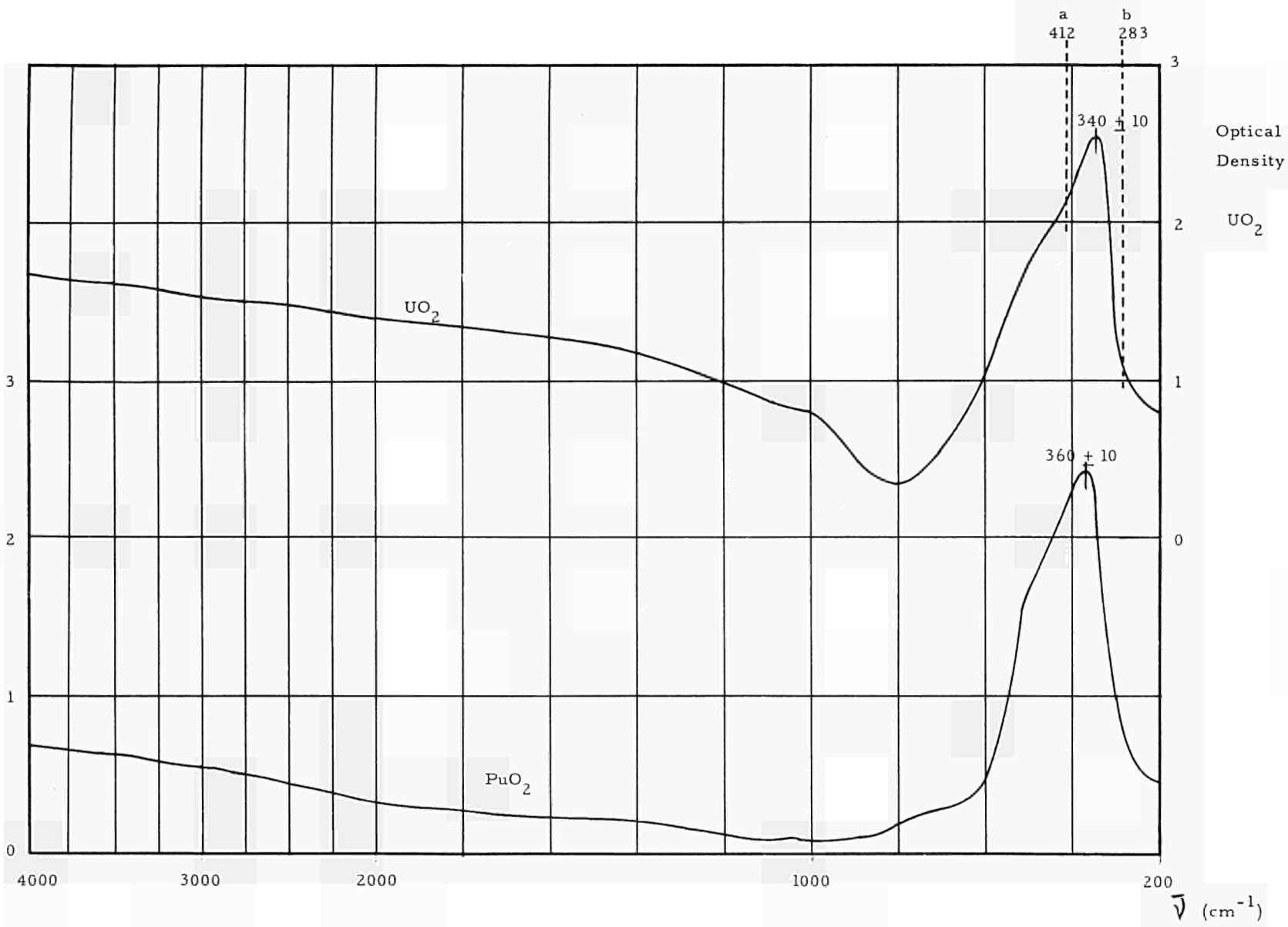

Fig. 3

Infrared absorption spectra of PuO_2 and UO_2 dispersed in Cs Br

a) Tsuboi et al., J. Chem. Phys., vol. 36 Nr. 5 (1301) march 1962

b) J. D. Axe et al., AED-Conf. 1966, 050 - 31

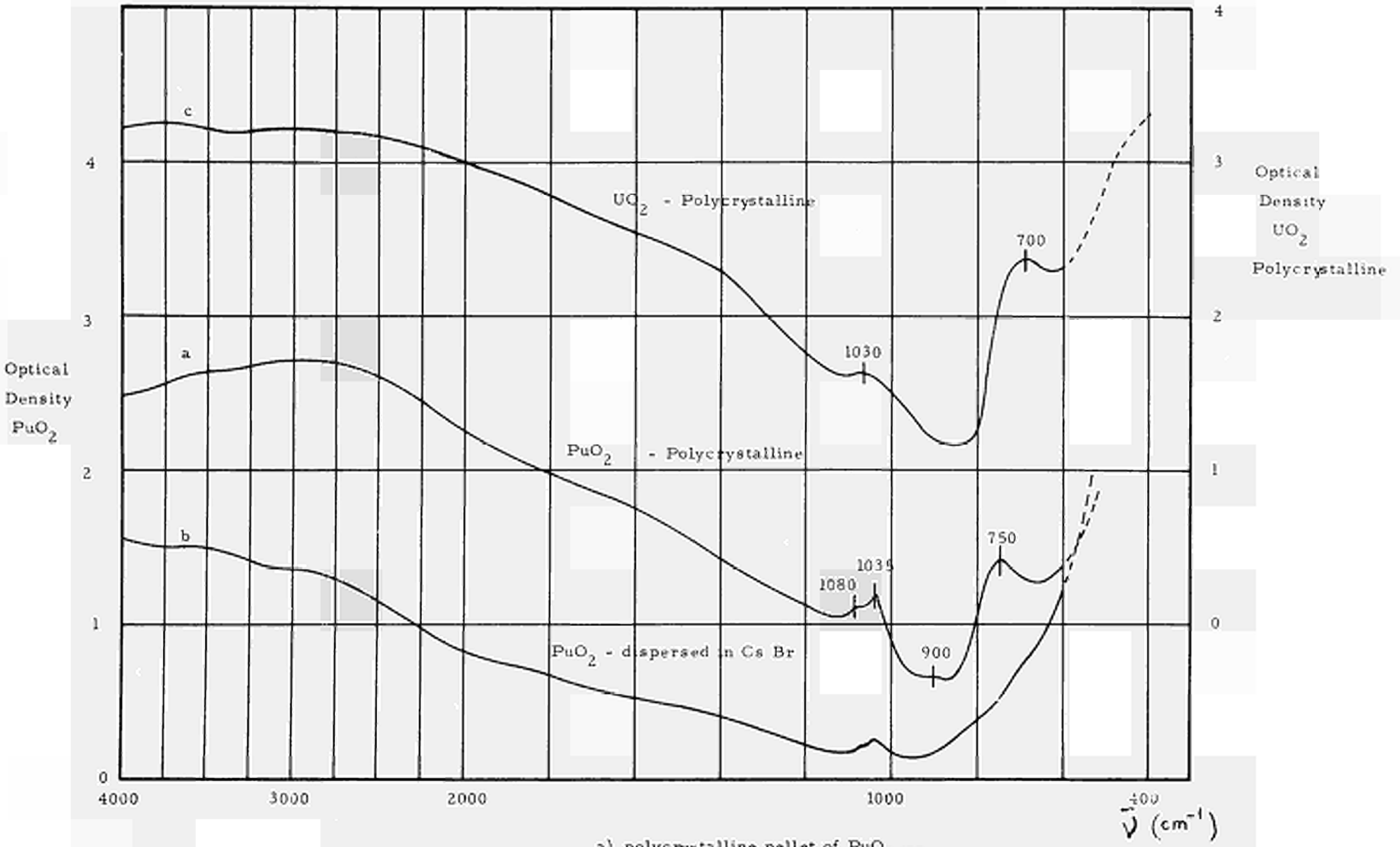

Fig. 4 Absorption spectra of

a) polycrystalline pellet of $PuO_{2.00}$
b) Cs Br - dispersed pellet (2% PuO_2 in Cs Br)
c) polycrystalline pellet of $UO_{2.00}$

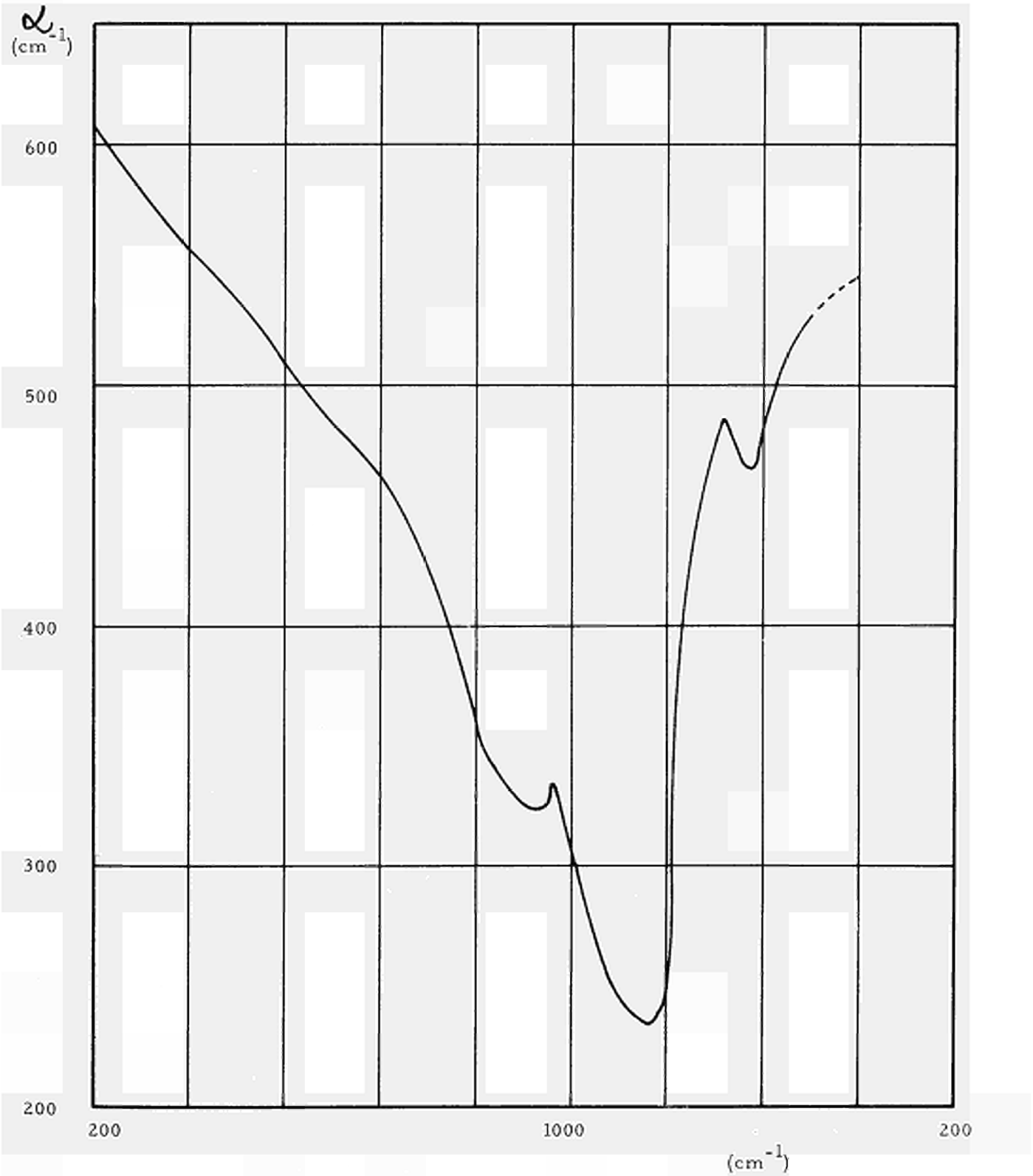

Fig. 5 Absorption coefficient of a polycrystalline sample of $UO_{2.00}$

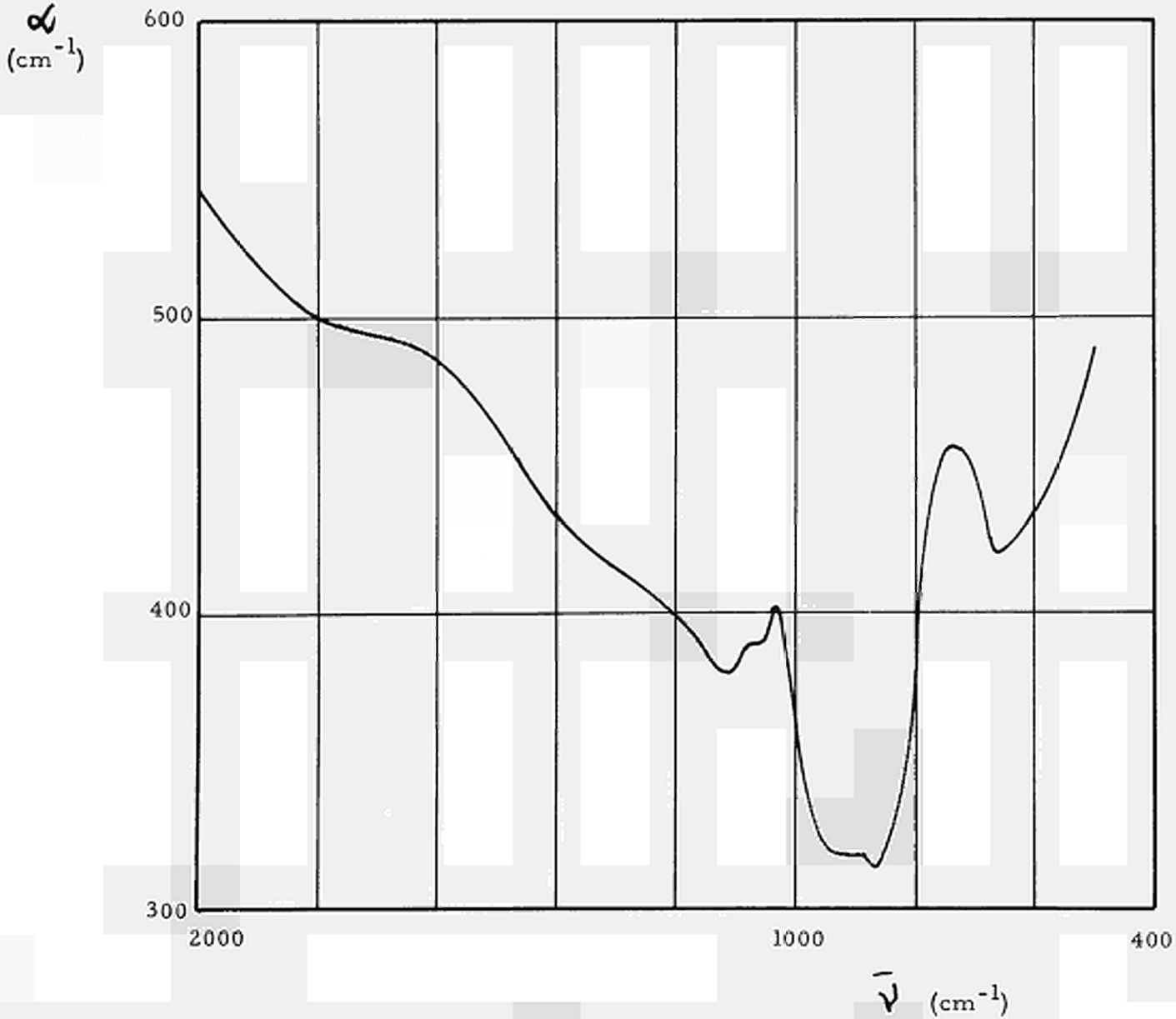

Fig. 6 Absorption coefficient of a polycrystalline sample of $PuO_{2.00}$

DISCUSSION

1. Properties of PuO_2

PuO_2 crystallises in the fluorite structure, space group O_h^5, and is isomorphous with both UO_2 and ThO_2. Its structure can be described as consisting of three interpenetrating f.c.c. lattices, a metal ion being at the origin, and the oxygen ions at $(\frac{1}{4}, \frac{1}{4}, \frac{1}{4})a$ and $(-\frac{1}{4}, -\frac{1}{4}, -\frac{1}{4})a$, where a is the cubic unit cell side. For PuO_2, a = 5,4960 Å at room temperature (for UO_2, a = 5,4682 Å, for ThO_2, a = 5,6066 Å).

2. One-phonon "Reststrahlen" absorption on infinite crystals

Table 1 gives a summary of the results. As already shown in fig. 3, the spectrum consists essentially of a single broad peak of high intensity. In the same table, a comparison is made with previously reported measurements on UO_2 and ThO_2. One may remark a discrepancy between the values obtained with polycrystalline materials and those obtained on a single crystal. This discrepancy will be discussed in the following section.

That a single line may be expected in the fluorite structure, can be concluded from the following considerations.

Since there are three atoms in the primitive unit cell, there will be in general nine branches of the dispersion relations in any direction. We assume that the crystal obeys Born-Karman boundary conditions. For the long wave length limit of vibration (i.e. at the Γ point of the Brillouin zone) group theoretical treatment shows that the nine possible degrees of freedom must form a basis for the irreducible representation $2\Gamma_{15} + \Gamma_{25}'$ (in Schoenfliess symbols) (6). One triply degenerate Γ_{15} mode constitutes the acoustic vibration of the ionic crystal. The Γ_{25}' mode is the Raman active mode, which is infrared inactive. In the first order, only the remaining Γ_{15} mode is infrared active. It is well known that, due to the macroscopic field associated with its longitudinal component, its triple degeneracy is lifted, so that it splits into

TABLE 1 : Values of the main resonance frequency ν (cm^{-1}) for ThO_2, UO_2, PuO_2

		ThO_2	UO_2	PuO_2
This work	Absorption measurements on powders dispersed in Cs Br	-	340 ± 10	360 ± 10
Tsuboi et al. (4)	Absorption measurements on powders dispersed in polyethylene films	-	412	-
J. D. Axe et al. (1)	Reflection measurements on single crystals	x) a) 279 ± 2 b) 282, 7	x) a) 278 ± 2 b) 283, 2	-
Dolling et al. (5)	Neutron scattering measurements on single crystals	-	284 ± 4	-

x)
a - Kramers - Kronig analysis
b - Classical dispersion analysis

a longitudinal and a transverse vibration. The longitudinal vibration has, for k = 0, a higher frequency, usually referred to as ω_{LO}, and is also infrared inactive. The transverse vibration is the only one infrared active, and its frequency is usually referred to as ω_{TO}. In the actual lattice it corresponds to a motion in which the two oxygen ions move together and the metal ion moves in the opposite sense.

Table 1 shows also that ThO_2 and UO_2 (when measured on single crystals) and PuO_2 and UO_2 (in our results) give rise to a similar resonance frequency. Although the discrepancy between single crystals and polycrystalline measurements remains, one can affirm that the behaviour of the different oxydes, when measured in the same conditions, is very similar. This is not a very striking fact, if one takes into account their isomorphism.

3. Finite crystals considerations

The discrepancy for UO_2 in the values measured on single crystals and on powders has already been remarked. We want now to discuss the considerations made in the introduction of this report. In powders, crystals are usually comparable with or smaller than the wave length of infrared radiation. This is certainly the case with our powdered samples, in which the greatest part of the crystallites have linear dimensions smaller than 10 μ.

In such a case, the resonance will occur at a frequency ω_o which is situated between the two limiting frequencies ω_{TO} and ω_{LO}, defined above for an infinite crystal. The values of ω_o will be furthermore dependent on the sizes and shapes of the particles. In fact, due to the existence of long range dipolar forces determining the dynamical behaviour of the lattice, in the case of finite crystals it is no more possible to distinguish purely transverse and purely longitudinal vibrations in the polarisation waves of the lattice. This very interesting phenomenon has been treated theoretically by many authors (1), (7), (8), (9).

As a consequence, all the physical properties depending on the vibrational spectrum of the crystal are affected by the size and the shape of the crystal.

The resonance frequency ω_o for a model consisting of a small sphere having a diameter smaller than the radiation wave length has been calculated explicitly by all the authors mentioned above, starting from the properties of an infinite crystal. Such a model obviously constitutes an oversimplification, because in a real situation one is confronted with a distribution of various sizes and shapes. This distribution will affect both the position of the maximum and the band width of the observed line. However, even if it is impossible to interpret rigorously the observed band using that model, one may be attempted to apply it on the assumption that the value of the maximum is the true ω_o for the largest portion of the particles, and that this portion is constituted mostly by small particles of a somewhat spherical shape. Evidence of the second assumption has been provided in our case by microscopic examination of the powders.

Table 2 shows, for UO_2, a comparison between AXE's measured values of ω_{TO} for a single crystal, and ω_{TO}'s calculated, for powders, by means of the equation

$$\omega_o^2 = \frac{\varepsilon_o + 2\varepsilon_M}{\varepsilon_\infty + 2\varepsilon_M}\ \omega_{TO}^2$$

derived by J.D. AXE (1) for this model. Here ω_o is the experimental resonance maximum on powders, ε_o, ε_∞ are respectively the static and the high-frequency dielectric constants, ε_M is the dielectric constant of the matrix in which the small spheres are embedded. A similar comparison is made for the compressibilities of UO_2, which are likewise calculated from the ω_{TO}'s by means of a well known equation relating ω_{TO}, ω_R (Raman frequency) ε_o, ε_∞ with the compressibility (see J.D. AXE (1)). Values of ε_o, ε_∞, ω_R for UO_2 are taken from the same paper of J.D. AXE.

Table 2: ω_{TO} and compressibility ß calculated for powders by J.D. AXE's equation (1)

	this work	TSUBOI	J.D. AXE
$\omega_{TO}(cm^{-1})$	250 - 265	250 - 265	278 ± 2 (KKA)[x]
ß (dyne cm^{-2})			283 ± 2 (CDA)[xx]
$x10^{12}$	0,58 - 0,62	0,58 - 0,62	0,478
meas. value: 0,472 (1)			

[x] KRAMERS - KRONIG analysis [xx] Classical dispersion analysis

The agreement is not very satisfying, but one may get from this table an idea on the type of information that may be derived from measurements on powdered samples in the actual state of the theory. One must conclude that our experiments give a qualitative support to the finite crystals theory. This is of some importance because not much experimental evidence is available on this respect [see (9) and (10) about the case of thin layers].

One may add that a displacement of the maximum to about 305 cm^{-1} occurred in some experiments run with UO_2 sintered powders, where the sizes of the crystallites were certainly 3 - 4 times bigger than in the other powders. This fact is also qualitatively consistent with the above reported theory.

4. Short wave length tail of the "Reststrahlen" band

Fig. 4 shows the short wave length side of the "Reststrahlen" band as measured on sintered pellets and on a very concentrated CsBr-dispersed sample. Fig. 7 shows a comparison of absorption spectra on ThO_2, UO_2 and PuO_2 polycrystalline samples. For UO_2 and ThO_2 absorption measurements on single crystals have been reported (1), (11). For UO_2 they agree, in the position of the peaks, with those found for polycrystalline materials.

For ThO_2 an additional peak has been detected at 8,5 μ. One may remark the similarity of the spectra.

No comprehensive interpretation has been reported of UO_2 and ThO_2 spectra. This is due mainly to a lack of additional information about the vibrational structure of the two crystals. Nevertheless, all the bands appear in the two- and three-phonons region of the spectrum, and they may be tentatively explained as combinations of fundamental vibrational modes.

Selection rules for the two-phonon combinations in the fluorite structure have been worked out by GARNESAN and BURSTEIN (6). One may see in this paper that a considerable number of combinations is possible, much greater than the actual number really found in the absorption spectra of these oxides.

The similarity of the spectra shown in fig. 7 points to similar vibrational properties for the three oxides, and to some fundamental reason for the appearance of only a few of the theoretically predicted combinations.

Starting from DOLLING's (5) neutron scattering data for UO_2, J.D. AXE (1) interprets the peak of UO_2 at 9,6 - 9,8 μ as a combination of the modes of the Raman and the longitudinal branches at the surface of the Brillouin zone. In view of the similarity of the spectra, possibly an equivalent explanation applies for the analogous peaks in ThO_2 and in PuO_2.

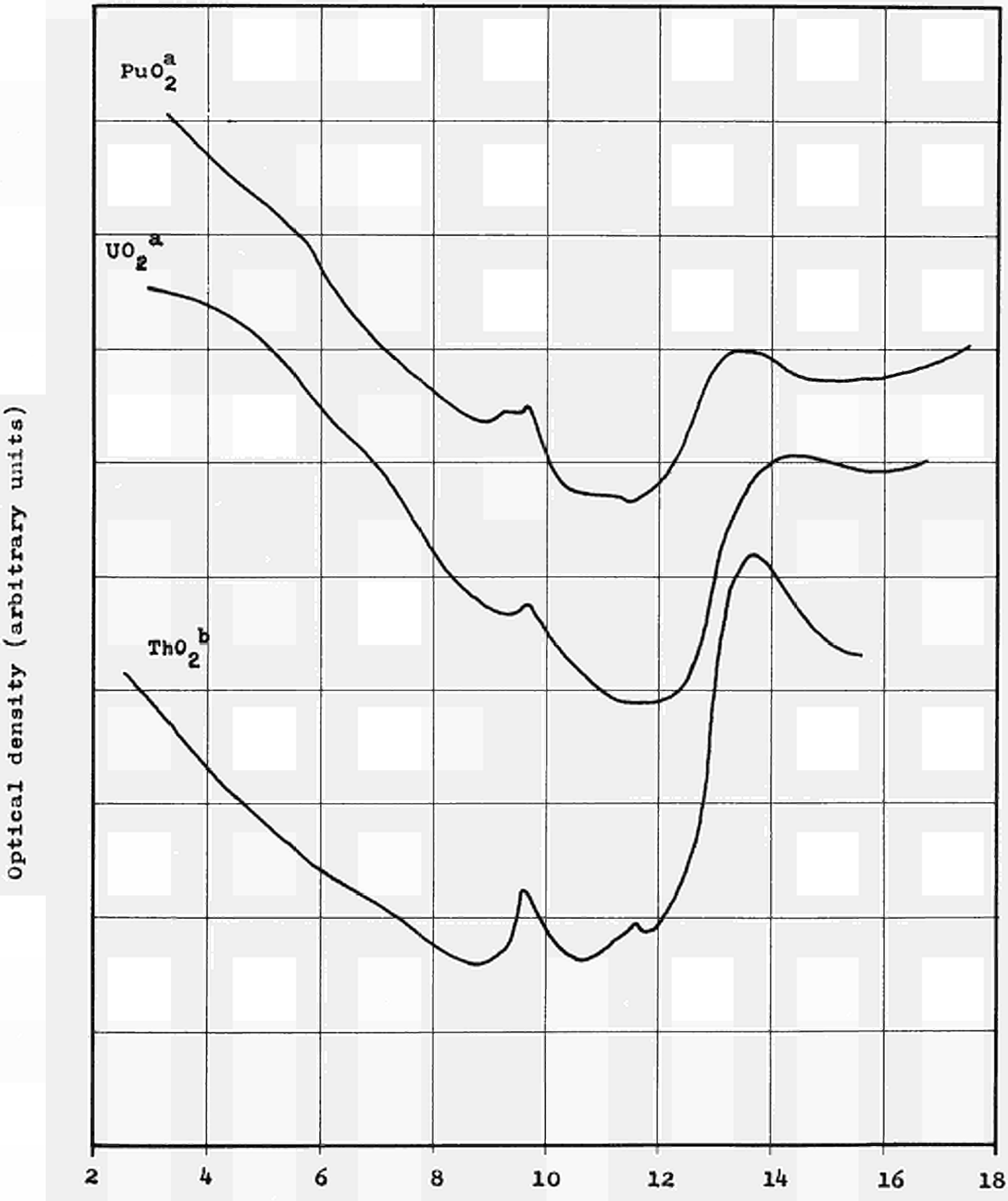

Fig. 7 Infrared absorption spectra of ThO_2, UO_2, PuO_2 sintered pellets.

a) Our measurements

b) J.L. Bates, see ref. (11) .

CONCLUSION

The infrared spectrum of PuO_2 bears a great similarity to the spectra of UO_2 and ThO_2. It was confirmed that the fundamental vibrational frequency depends on the size and shape of the crystallites studied.

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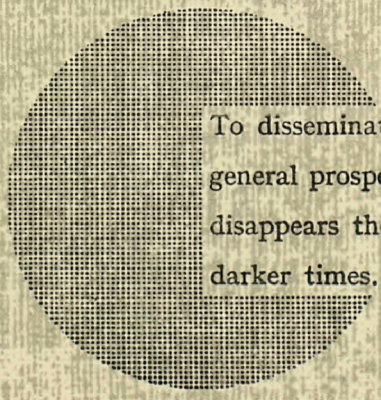

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Alfred Nobel

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