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Optical properties of Xe under very high pressure (*) (+)

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Résumé. — Le xénon a été étudié, dans l'enclume diamant jusqu'à 63 GPa. Aucune transition vers un état métallique n'a été observée, ce qui contredit certains résultats antérieurs. La limite d'absorption observée montre qu'il a encore dans ce domaine de pressions, une bande interdite de plusieurs eV. Le croisement des bandes ne doit pas se produire, dans le xénon au-dessous de 100 GPa (1 Mbar) si la structure c.f.c. reste stable.

Abstract. — Xenon has been studied in the diamond anvil cell up to 63 GPa. It does not exhibit any insulator → metal transition, contrary to previous reports. The absorption observed shows that its bandgap is still several electron-volts in this range of pressures. Band closing should not occur in Xe before 100 GPa (1 Mbar) if the f.c.c. structure remains stable.

1. **Introduction.** — Recent developments in static ultra-high pressure methods (diamond anvil cells) as well as improvements in dynamic techniques, have led to numerous observations of insulator-metal phase transitions in elementary systems. Among these, rare gas solids are by far the easiest to handle from a theoretical point of view since they are monoatomic and have a complete electronic shell. In this series, the calculated dielectric-conductor transition pressures decrease with increasing atomic number and therefore, xenon is the first candidate for the observation of metallization in rare gas solids. Calculated transition pressures for this element range between 40 and over 200 GPa [1-4].

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Under pressure, the indirect $\Gamma_{15}^V X_1^C$ edge decreases below the direct $\Gamma_{15}^V \Gamma_1^C$ gap and when it goes down to zero, xenon should exhibit metallic, or rather semimetallic behaviour. This is different from structural phase transitions to a metallic state, which are found in a number of insulators and semiconductors. Recent calculations, on the other hand [4], suggest that the low-pressure f.c.c. structure might go into b.c.c. under pressure; this transition might occur before band closing or even originate it.

Most static [5] and dynamic [1, 6] experiments confirm EOS calculations [1, 2] that predict band closing to occur above 100 GPa. On the other hand, a set of transport measurements [7] with the diamond indenter method, shows between 30 and 50 GPa a decrease of several orders of magnitude in the resistivity of the interdigitated electrodes when the setup is loaded with Xe. The authors of [7] interpret this observation as evidence for a transition to the metallic state.

In this letter, we give preliminary results on the optical properties of xenon under high pressure which show that it remains a dielectric even above 60 GPa. The electrical behaviour that has been reported [7] can be assigned neither to a transition to a metal nor to a small-gap semiconductor.

2. Experimental. — The mechanical part of the diamond anvil cell that was used has been described before [8]. This setup was modified to allow loading with xenon at 300 K under 50 bars (5 MPa). A removable attachment allows evacuation of the cell, including the experimental space, and loading with 4.5N xenon. Under the present conditions, a sample with comparatively large mass, over one microgram, can be retained between the diamonds. Type I white diamonds were used. Before loading, a ruby ($\text{Al}_2\text{O}_3 : \text{Cr}$) chip, some 30 microns across and 15 microns in thickness, is placed in the centre of the cell. Under 100 milliwatt irradiation from the 514.5 nm line of an Ar^+ laser, its luminescence wavelength shift serves as a pressure gauge. The pressure coefficient was taken to be constant and equal to $7.53 \text{ cm}^{-1} \cdot \text{GPa}^{-1}$.

Under pressure, the anvil parallelism remains sufficient to allow measurement, under a microscope, of the Fabry-Pérot interference spectrum, with a double Jarrell-Ash monochromator. The optical path in xenon ne (n : refractive index; e : the thickness of the sample) can be derived from the interfringe wavenumber $\Delta\nu$, neglecting the dispersive term, by :

$$2 ne \Delta\nu = 1.$$

The refractive index of Xe under pressure could be evaluated at two pressures :

i) At 6 GPa, index matching of Al_2O_3 (ruby) and Xe was observed (the ruby chip « disappears » in the field of the microscope). After corrections for the pressure variation of the index of refraction of ruby, this yields a value of $n = 1.75 \pm 0.02$, the index anisotropy being neglected.

ii) Around 40 GPa, the amplitude of transmission interference extrema goes to zero when the index of Xe matches that of diamond. The estimated value is 2.39 ± 0.03 . These values, together with the low-density data, are sufficient to draw the $n(P)$ curve. From experimental values of $2 ne = 1/\Delta\nu$, the thickness of the cell *versus* pressure can be evaluated. This curve is shown on figure 1. It is extrapolated above 30 GPa where interferences are weak.

Optical transmittancy was measured under a microscope in the spectral range 1.5 to 3 eV. Total light illumination was provided by a halogen burner with a regulated (10^{-3}) power supply. Optical observation of Xe without spectral measurements was done in a first set of experiments up to 63 GPa. In a second run, optical transmittancy was measured and anvil failure occurred slightly above 53 GPa. In our setup, no reference beam is directly available for absolute intensity measurements. Nevertheless it was verified that the optical response of the setup (source; microscope; anvils; sample; monochromator; photomultiplier) remains constant at all pressures below 30 GPa.

This response was taken as the zero absorbancy standard to determine the absorption of the xenon sample and diamond anvils at pressures above 40 GPa where an absorption edge appears in the high-energy region of the spectrum.

Fig. 1. — Thickness e of the Xe sample between the anvils *versus* pressure. Full line : mean value. Dashed lines : upper and lower values for the fit.

Fig. 2. — Absorption of the xenon sample and the diamond anvils. Experimental points (stars) are computed with the thickness extrapolated from figure 1, assuming zero absorption from the diamond anvils. The full line is a fit with a quadratic law corresponding to an indirect absorption process in xenon (see text).

3. Results and discussion. — Optical observation under the microscope shows that xenon remains transparent in the visible up to 63 ± 2 GPa. No discontinuity in the grain boundaries of the sample could be observed. The absorption spectrum given in figure 2, at 53 GPa, is given under the assumption that the anvils do not introduce any extra absorption under high-pressure and that the observed spectrum is solely due to the Xe sample, the thickness of which is given by

the diagram in figure 1. The full line is a least-square fit with a quadratic law for an indirect $\Gamma_{15}^V-X_7^C$ transition, the energy of the phonons being neglected. The value we obtain is 2.3 eV for the gap; although this closely fits with calculations by M. Ross and A. K. McMahan [1] on the basis of a Hedin-Lundqvist potential, this might be coincidental and this value may be only a *minimum* value: Type I diamonds do exhibit a weak band above 3 eV and a continuum at 4 eV both due to nitrogen which join with the intrinsic edge above 5 eV. No data exist on the behaviour of those impurity levels under pressure and this edge may well come from the anvil material. This coincidence with calculations with a H.L. potential is unexpected since a more realistic Slater-type potential [1] gives values of the order of 4 eV, in the gap, in this pressure range.

Recent optical results [9] up to 43 GPa support this last value. In any case, in the high-energy part of the spectrum, xenon behaves like a broad-band ($E_g \geq 2.3$ eV) semiconductor or insulator. The behaviour of the refractive index provides qualitative confirmation for this, up to 30 GPa where it could be measured:

The interfringe *variation* with photon energy, which is representative of index dispersion, remains the same to within 3 % between 0 and 30 GPa. This would not be the case if a high-energy transition had come down close to the visible region under high pressure. On the low-energy side, no near-infrared absorption on free carriers was observed down to 1.5 eV.

4. Conclusion. — The results of this work may be summarized as follows:

- Xenon remains optically transparent above 60 GPa.
- The refractive index increases with density from 1.4 to more than 2.4 at 50 GPa. Its dispersion with wavelength, on the other hand, does not vary up to 30 GPa.
- The optical gap at 53 GPa is 2.3 eV or possibly more.
- These observations contradict previous conclusions [3, 7] on the occurrence of a transition to a metallic state between 30 and 50 GPa. Electrical measurements [7] in that pressure range were done at 32 K whereas our data were collected at 300 K, but this difference in temperature does not help reconcile the results: the bandgap width is, to first order, a density effect and the temperature correction to density between 0 and 300 K is negligibly small as compared to its variation [1] between 30 and 60 GPa. The decrease in resistance observed in reference [7] can be assigned neither to a first order transition to a metallic state nor to continuous closing of the gap.
- Finally, even if the bandgap is as small as shown in figure 2, comparison with existing EOS and bandgap calculations [1, 2] still places band closing above 100 GPa (1 Mbar).

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