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LA-UR-88-2559

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DE88 016230

TITLE VERY HIGH PRESSURE MOSSBAUER SPECTROSCOPY USING DIAMOND ANVIL CELLS

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SUBMITTED TO Invited paper, Third Seeheim Workshop on Mossbauer Spectroscopy, May 25-27, 1988, Seeheim, FRG

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VERY HIGH PRESSURE MÖSSBAUER SPECTROSCOPY USING DIAMOND ANVIL CELLS

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The technique of generating very high pressure by means of Diamond Anvil Cells (DAC) for Mössbauer Effect applications is outlined. A comprehensive description is presented of the principles of DAC, modification for the use in Mössbauer Spectroscopy (MS), the Merrill-Bassett and Bassett cells, of pressure measurements, of gasketing and collimation, and of hydrostatic media. Examples of ^{151}Eu, ^{119}Sn and ^{129}I are given showing the feasibility of DAC applications in MS. Other isotopes with potential use for high pressure MS using DAC are suggested.

PACS numbers: 62.50.+p, 31.70.Ks, 61.16.-d, 76.80.+y

1. INTRODUCTION

Static high pressure techniques have been widely applied since the early days of the Mössbauer Effect to investigate the effect of lattice spacing changes on the hyperfine interaction constants. The devices to achieve high pressure was based primarily on the Bridgman anvil and the piston-cylinder concepts, both of which were invented and developed by Bridgman and later improved by Drickamer and co-workers /1,2/. Reviews of Mössbauer Spectroscopy with high pressure based on Bridgman cells were written by several authors during the sixties /3,4,5,6/, and, at the end of the seventies by Williamson /7/, who emphasized the role of high pressure on isomer shifts On the practical side, the use of Bridgman cells for Mössbauer Effect was limited to ~150 Kbar maximum pressure /8/.

During the last 15 years, concurrently with numerous high pressure studies with the Bridgman cells, a new technique was evolved which revolutionized the field of static high pressure, namely the <u>diamond anvil cell</u> (DAC). Today the DAC has become the most powerful ultra-high-pressure device, opening new venues for exciting studies of condensed matter. The first Mössbauer Effect DAC studies were reported as early as 1976 by Huggins, Mao, and Virgo /9/ using an iron absorber. Source experiments with ^{57}Co(Fe) were first reported by Taylor and co-workers in 1982 /10,11/. Other MS experiments with ^{57}Fe (iron metal and Fe_2O_3) to pressures beyond 500 Kbars were conducted by Syono et. al. /12/ and Nasu et. al. /13/. The first ^{119}Sn experiments in tin metal with DAC were reported by Chow et.al /14/, ^{129}I in elemental I_2 by Pasternak et.al., in 1986 /15/, and ^{151}Eu in Eu metal by Farrell and Taylor /16/. Those studies proved that DACs can practically be used as an ultra-high-pressure device not only for ^{57}Fe MS, but also for other Mössbauer isotopes providing the experimental conditions are properly optimized.

The purpose of this paper is to introduce the DAC device technique for generating high and ultra-high pressure in conjuction with the Mössbauer Effect. It will be shown that by proper modification of the existing DACs it is possible to conduct MS with absorbers with pressures at least to 1 Mbar. As an example for this application some recent results will be presented /17/ of ^{119}Sn and ^{129}I MS in SnI_4, a molecular crystal showing unusual structural and valence transformations at elevated pressure, and of ^{151}Eu in Europium metal /16/. We shall concentrate on the principles of the DAC, emphasizing: 1- construction of the very simple modified Merrill-Bassett cell /18/ and the more sophisticated Bassett cell /19/, where pressures to 350 Kbar and beyond 1 Mbar are achieved, respectively; 2- in-situ pressure calibration and, 3- particular details such as γ-ray collimation, sources, hydrostatic pressure media, etc. For a general review of the DAC the reader is referred to the paper of A. Jayaraman /20/.

2. TECHNIQUES

2.1 Principles of Diamond Anvil Cells

The DAC is formed from three basic parts, namely: 1- a force generating device, 2- diamond anvils, and 3- a sample chamber made from a metal gasket. The schematic drawing describing the basic principle of the DAC is shown in Fig.1. The high pressure achieved with the diamond anvils is due to the extremely high strength of the diamond single crystals and the relatively small size of the sample chamber which allows use of relatively small forces achieved normally by the use of high strength steel screws.

Diamond anvils are made from high quality, flawless, brilliant cut gem stones. For most high pressure applications diamonds in the range of 1/8 to 1/3 carat are used. The culet tip is ground and polished to a hexa decagon surface parallel to the table in the (100) or (110) plane of the diamond (see Fig.1). The diameter of the anvil is in the range of 0.5 - 1.0 mm. For pressures exceeding 1 Mbar a bevel of 2 - 7° is added to the anvil face, producing a smaller anvil surface of ≈0.2 mm. Diamond anvils are commercially available either in standard or custom-made configurations /21/.

The gasket and the sample chamber are important factors in generating high and hydrostatic pressures with DACs. The sample chamber is prepared by drilling a hole (0.2 - 0.3 mm in diameter) at the center of an indentation made by the anvils. The final thickness of the compressed gasket is in the range of 20 - 40 μm. The gasket apart from providing containment of the pressure medium acts as a supporting ring around the diamond tip, preventing failure of the anvils due stresses at the edge of the anvil faces.

2.2 Pressure Calibration

Due to its transparency to light one is able to use the ruby fluorescence method to measure in situ pressures and pressure distributions. The two R lines of ruby, R_1 and R_2, are quite intense, fluorescing at 692.7 and 694.2 nm, respectively. They can be locally excited, for example by the green or blue lines of an argon ion laser or by the blue line of a Cd-He laser. The wavelength of the R-lines increase with pressure, linear to at least 300 Kbar with the simple relation: $P = 27.46\Delta\lambda$ where P is in Kbar and $\Delta\lambda$ in nm. The following relation holds to 1 Mbar /22/:

$$P(\mathrm{Mbar}) = 3.808[(\Delta\lambda/694.2 + 1)^5 - 1] \qquad (1)$$

where $\Delta\lambda$ is nanometers. Ruby chips in the 10 - 50 μm size range are spread below and on top of the sample. A laser microscope system for pressure calibration is shown in Fig.2 /23/. It is composed of a high quality metallurgical microscope, a dichroic mirror to allow the blue(green) light of the exciting laser to be almost 100% reflected into the DAC and the red fluorescent ruby light to be transmitted, a fiber optic bundle to transmit the light into a monochromator, and a 0.1nm resolution monochromator. The laser-calibration system is a very important part of the DAC high pressure research. Pressure calibration using several ruby chips allows measurement of the pressure distribution at a given setup /13/. One should be aware of the differences in pressure while measuring the ruby fluorescence at room temperature and the Mössbauer spectrum at low or high temperatures. In the case of the cell used by us a 5% pressure difference was detected between 20 K to 300 K. The variation of pressure with temperature is a characteristic of the particular DAC used and should be experimentally determined.

2.3 Pressure Media

To obtain a hydrostatic medium for pressures to 150 Kbar a mixture of methanol-ethanol (4:1) is commonly used /24/. For higher pressures, condensed gases such as N_2, Ar or Xe are preferable. They are introduced into the sample chamber either by pressurizing the gas or immersing the cell into the condensed gas prior to sealing the cell.

In what follows we describe two well known types of DACs modified to account for the special requirements of the MS. In general they are manufactured from special steels achieving high degree of strength and hardness after proper heat treatment. A common non ferrous material used is a Cu-Be alloy, whose advantage is its non-magnetic property.

2.4 The Modified Merrill Bassett (MB) Cell

The MB cell is the simplest DAC. It is extremely compact and was designed by Merrill and Bassett for the purpose of studying x ray diffraction. At its inception it was meant to generate pressures up to 150 Kbars. However, in a recent application of this cell to ^{129}I MS /25/, pressures to 350 Kbar were achieved, and it is believed by the authors capable of pressures to 500 Kbar. A description of a MB cell showing its relative dimensions is shown in Fig.3. A photograph of a MB cell used by the authors is depicted in Fig.4. Such a cell has been used for ^{119}Sn /17/, ^{129}I /25/, and ^{151}Eu /16/ absorption experiments and for ^{57}Co(Fe) source studies /11,12/. For the absorption studies, a $Ta_{90}W_{10}$ alloy gasket was used. This high-Z material not only serves as a gasket to contain the sample but also as a collimator for the 21 - 28-keV γ rays pertaining to these isotopes. This material has excellent gasketing properties, it extrudes very well around the diamond and is believed to be the main factor for obtainig pressures to 350 Kbar in the MB cell.

The MB cell is composed of the following parts: 1- two opposite platens of hardened metals or alloys. Three screws are used to pull the platens together and provide the force for obtaining the high pressure. 2- two discs for holding the anvils. One of the discs can be horizontally moved for aligning the anvils. 3- anvils and, 4- gaskets. Parallel conditions of the two culets are achieved with the three screws normally used for generating the high pressure, by observing the interference pattern with the gasket removed. Any differences in the overall thickness measured at the three corners are maintained at all subsequent pressures.

An important aspect of the MB cell for MS is its adaptability to high magnetic fields and very low temperatures. Due to its small size and the possible construction using non-ferrous materials, it can be easily inserted in a standard large bore superconducting magnet or attached to a dilution refrigerator /10,11/.

2.5 The Bassett Cell

The "Bassett" cell was introduced by Bassett et.al /19/. A modified Bassett cell designed for MS is shown in Fig.5. With such a cell, one may reach 1 Mbar pressure using flat culets, and pressures exceeding 1 Mbar with bevelled culets . A version of this Bassett cell similar to one shown in Fig.5 was used by Syono et.al./12/ and recently by Masu et. al. /13/.

2.6 Special Considerations

For absorption measurements one should consider the problem of absorber sample sizes. This is an unique case where the absorber diameter is considerably smaller than that of the source! A typical absorber dimension is 300-μm diameter ($7x10^{-4}$ cm^2) and 30 μm thickness Due to its small weight, loading of absorbers done under a microscope with special tools. All commercial sources have areas far larger then the absorbers. It is desirable to reduce source diameters and at the same time to increase its specific activity. For instance in the case of ^{129}I Mössbauer Effect , a special Mg_3TeO_6 source was made in a form of 3-mm diameter pellet /26/. However successful experiments were conducted with commercial 5-mCi $CaSnO_3$ and 160-mCi $^{151}SmF_3$ sources. It is worth mentioning that with such large ratio of source/absorber radii, the absorber material close to the gasket periphery is mostly shadowed. Rough calculations show that the effective absorber diameter is 70% of the actual one. This reduction of absorber diameter has a positive aspect in the sense that the material close to the gasket (where the pressure inhomogenuity is the largest) is not effectively scanned. Thus, the absorption spectra emphasize those parts of samples with the least pressure inhomogeneity.

One important consideration pertaining to the Mössbauer Effect is the anvil thickness. An average thickness for a pair of 0.30-carat anvils is 4.5 mm. The electronic absorption of ^{151}Eu, ^{119}Sn, or ^{129}I γ-rays is around 0.50 whereas for ^{57}Fe is 0.93! Thus, for ^{57}Fe DAC experiments one should consider using smaller anvils, around 0.10 carat each, specially cut to increase the table size and decrease its thickness /27/.

3. EXAMPLES OF HIGH PRESSURE ^{119}Sn, ^{151}Eu, AND ^{129}I MÖSSBAUER SPECTROSCOPY

As an example of DAC applications for Mössbauer Effect studies we present some recent results of high pressure MS of ^{119}Sn /17/, ^{151}Eu /16/, and ^{129}I /[illegible]/ . These studies were performed in the temperature range of 4 - 300 K and with pressures up to 360 Kbar using the modified Merrill-Bassett cell as described in 2.4. Measurements were conducted in a bath-cryostat using He or N_2 exchange gas and with a source vibrated in a vertical mode. A 1-mm thick NaI(Tl) counter was used for most cases.

Mössbauer spectra of Eu metal at 44 K with varying pressure is shown in Fig.6. The source used was a 4-mm diameter 160-mCi SmF_3 at 44 K. Typical accumulation time for spectra was 24 - 48 hours. A sample 50- µm thick was placed in a 300-µm diameter hole drilled in a $Ta_{90}W_{10}$ gasket. Liquid argon was used as the hydrostatic pressure medium. As can be seen, despite the relatively low count rate (5 - 20 c/s), spectra of sufficient quality could be accumulated in the time period. Similar conclusions can be derived for the case of ^{119}Sn. In Fig.7 typical spectra of ^{119}Sn in a SnI_4 absorber at various pressure and at 70 K are depicted. Measurements were done with a 6.6-mg cm^{-2}, absorber thickness, enriched with ^{119}Sn, placed in a 270-µm-diameter 30-µm-high cavity. A 25 µm Pd filer was used. The source was a commercial 5-mCi $CaSnO_3$. Time periods for collecting each spectrum were 12 - 24 hours with a characteristic count rate of 50 c/s.

Mössbauer spectra with ^{129}I require more stringent precautions as compared with the Eu and Sn experiments. The ^{129m}Te source decays primarily by high energy beta decay thus producing intense and energetic Bremstrahlung radiation, that is easily transmitted through the cell and the gasket. Furthermore, the 27.8-keV Mössbauer line is heavily "contaminated" with the K-x ray of Te. Both these events contribute immensely to the background. A $Mg_3{}^{129}TeO_6$ source was used in the form of a 3-mm diameter, 0.5-mm thick pellet irradiated in a high flux reactor for three weeks period and then annealed. A typical spectrum $^{129}I_2$ at 270 Kbar and 4 K is shown in Fig.8. The absorber thickness was 20 mg cm^{-2}, loaded into a 210-µm diameter hole. Due to the complex features of the quadrupole split spectrum, 2-3 days were required to accumulate sufficient counts to disentangle the several sites and hyperfine constants to a substantial degree of confidence. Spectra recorded with a $Sn^{129}I_4$ absorber at 4K with increasing (up arrow) and decreasing (down arrow) pressures are shown in Fig.9.

4. CONCLUSION

So far we have presented DAC high pressure MS for several isotopes, namely: ^{57}Fe, ^{119}Sn, ^{129}I, and ^{151}Eu. For other MS isotopes one should consider:

1- The use of "point" sources with high specific activity.

2- Problems related to the particular γ-ray energy transition.

The first problem is straightforward. The problem of γ-ray energy concerns both low and high energies. In the case of low-energy γ-ray, the limit is the diamond thickness. At the moment, for absorption experiments 14.4 keV seems to be the lower limit, even with smaller (e.g. thinner) diamonds /27/. For isotopes with $E_\gamma \geq 35$ keV external collimators are needed unless "point" sources with diameters less then ≈300 μm are available. The following isotopes have many of the right properties for DAC studies: ^{67}Zn, ^{155}Gd, ^{161}Dy, ^{170}Yb, ^{193}Ir, ^{197}Au, and ^{237}Np.

The application of Mössbauer Spectroscopy to very high pressures, using DACs has been established. Undoubtedly, modification of existing DACs and development of new concepts specifically addressing MS will continue. Consequently, we may expect a revival of high pressure MS studies extending to the very high, megabar region. The need for these studies is clear. The Mössbauer Effect is an unique "microscopic probe" operating on the atomic-molecular-scale, with many special features for measuring properties of condensed matter at very high pressures! In particular we envisage a *strong demand for this technique in geophysical and geochemical studies*. It is well known that iron is a prevailing element and a dominant part of most compounds forming the earth and its interior where pressures in the Mbar region are prevalent.

Furthermore, we may expect an increasing number of MS studies in materials that undergo phase transitions at very high pressures, supplementing x-ray diffraction and optical data.

5. ACKNOWLEDGEMENTS

This work was funded in parts by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Material Sciences and by the U.S.-Israel Binational Science Foundation under Grant No. 86-00241.

References.

/1/ H.G. Drickamer, in **Solid-State Physics**, Vol. 17, eds. F. Seitz and D. Turnbull (Academic Press, New York, 1965) p1.

/2/ H.G. Drickamer, op.cit.., Vol. 19 (1966) p135.

/3/ H. Frauenfelder and R. Ingalls, in **Applications of the Mössbauer Effect in Chemistry and Solid State Physics**, (Technical Reports Series, IAEA, Vienna, 1966), No.50.

/4/ H.G. Drickamer, R.Ingalls, and C.J.Cotton, The Mössbauer Effect at High Pressures, in **Physics of Solids at High Pressures**, eds. C.T. Tomizuka and R.M. Emerick, (Academic Press, New York, 1965).

/5/ H.G.Drickamer, S.C.Fung, and C.K. Lewis, Jr.,High Pressure Mössbauer Studies in **Advances in High Pressure Research**, Vol. 3, ed. R.S. Bradley, (Academic Press, New York, 1969).

/6/ W.B. Holzapfel, High Temperatures - High Pressures 2(1970)241.

/7/ D.L. Williamson, in **Mössbauer Isomer Shifts**, eds. G.K. Shenoy and F.E. Wagner, (North Holland, Amsterdam, 1978) p317.

/8/ Pressure is expressed in Kbar (kilobar) = 10^3 bar and Mbar (megabar) = 10^6 bar. The unit currently used (MKS) is GPa (gigapascal) = 10^4 bar. 1 bar= 10^5 N m^{-2} = 0.9869 atm = 1.0197 kg cm^{-2}.

/9/ F.E. Huggins, H.K. Mao, and D. Virgo, in **Carnegie Institute of Washington Year Book**, 74(1975)405.

/10/ G. Cort, R.D. Taylor, and J.O. Willis, J. Appl. Phys. 53(1982)2064.

/11/ R.D. Taylor, G.Cort, and J.O. Willis, ibid., 8199.

/12/ Y. Syono, A. Ito, S. Morimoto, T. Suzuki, T. Yagi, and S. Akimoto, Solid State Comm. 50(1984)97.

/13/ S. Nasu, K. Kurimoto, S. Nagatomo, S. Endo, and F.E. Fujita, Hyp. Int. 29(1986)1583.

/14/ L. Chow, P.A. Deane, J.N. Farrell, P.A. Magill, and L.D. Roberts, Phys. Rev. B33(1986)3039.

/15/ M. Pasternak, J.N. Farrell, and R.D. Taylor, Hyp. Int. 28(1986)837.

/16/ J.N. Farrell and R.D. Taylor, Phys. Rev. Lett. 58(1987)2478.

/17/ M. Pasternak and R.D. Taylor, Phys. Rev. B37(1988)8130.

/18/ L. Merrill and W.A. Bassett, Rev. Sci. Instrum. 45(1974)290.

/19/ W.A. Bassett, T. Takahasi, and P.W. Stook, Rev. Sci. Instrum. 38(1967)37.

/20/ A. Jayaraman, Rev. Mod. Phys. 55(1983)65.

/21/ Diamond anvils are currently manufactured by D. Drucker, Amsterdam, and Ramot, Tel Aviv.

/22/ H.K. Mao and P.M. Bell, Science 200(1978)1145.

/23/ Variations of the scheme shown in Fig.2 are used at many high pressure laboratories (see P.M. Bell and H.K. Mao in **Carnegie Institution of Washington Year Book** 74(1975)399). The version shown is being used at Tel Aviv University.

/24/ G.J.S. Piermarini, S. Block, and J.S. Barnett, J. Appl.Phys. 44(1973)5377.

/25/ M. Pasternak, J.N. Farrell, and R.D. Taylor, Phys.Rev.Lett. 58(1987)575.

/26/ M. Pasternak, M. Van der Heyden, and G. Langouche, Nucl. Instrum. Methods, Phys.

Res. Sect. B4(1984)152.

/27/ Special 0.09 - 0.10 carat anvils for ^{57}Fe MS, with increased table size and a total thickness of the anvil pair of 2.4 mm, are currently produced by Ramot, P.O.Box 39296, Tel Aviv 61392, Israel.

Figure Captions.

Fig.1 The basic opposed diamond anvil configuration used for DACs to pressures of $\leq$1 Mbar. For pressures exceeding 1 Mbar an additional bevelling is required (see text). Ruby chips are spread at the sample top and bottom for pressure measurments.

Fig.2 Schematic of a laser-microscope. 1) A BHT Olympus microscope, (2) Illuminator, (3) x - y stage, (4) DAC, (5) An extra long-working-length 20X objective, (6) Illuminator, (7) A 20 - 160 mW variable power argon-ion laser. (8) Laser power controller. (9) A dichroic mirror passive to red and reflecting for the blue (green) light. (10) Filters. A combination of filters and the blue or green options of the dichroic filter (9) allows the use of the blue+green, blue or green modes of the laser. (11) Occular. (12) Fiber optic bundle with a circular cross section at the microscope exit and a rectangular section matching the entry slit of the monochromator. (13) Monochromator (0.1 nm resolution) with a computer controlled step-motor. (14) 12mm-diameter photomultipier. (15) PC for controlling the monochromator and collecting and analyzing data from the photomultiplier. (16) Camera.

Fig.3 A cross-section of the modified MB-DAC. 1 Platens with three steel screws. 2 Supporting disks. 3 Ta$_{90}$W$_{10}$ gasket/collimator. 4 Diamond anvils. 5-Vibrating source. 6- Compressed gasket with the absorber chamber. 7- Ruby chips. 8 Absorber.

Fig.4 A photograph of copper beryllium Merrill Bassett DAC. The upper part shows the assembled cell. On the left of the lower part one notices the gasket (held by two pins), and the indented part where the absorber chamber is drilled. The diamond anvil glued to the disk is shown at the right part.

Fig.5 A sketch of a modified Bassett cell used for MS. The anvils are placed between the upper (1) and lower (2) tungsten carbide rockers. While adjusting the anvils, the rockers can be freely rotated (x_r and y_r), whereas the translational alignments (x_t and y_t) are achieved using external fine screws. 3- Cylinder. 4- Piston. 5- Thrust ring. 6- Six screws. 7- Vibrating source.

Fig.6 Mössbauer spectra of Eu metal at 44 K and 0, 62, and 98 Kbar. The source used was $^{151}SmF_3$ at 44 K.

Fig.7 Absorption spectra of $^{119}SnI_4$ at 140 K and various increasing pressures. The configurational phase transition of the Sn(IV) central ion with IS = 3.8 mm sec^{-1}, taking place with increasing pressure is clearly observed.

Fig.8 Mössbauer spectrum of $^{129}I_2$ at P = 270 Kbar and T = 4 K and with a Mg_3TeO_6 source at 4 K. The solid line is a least-squares fit to the experimental points assuming three quadrupole split components (dashed and dotted lines).

Fig.9 Mössbauer spectra of $Sn^{129}I_4$ for increasing (up arrow) and decreasing (down arrow) pressures at 4 K. Spectra above 90 Kbar were fitted with two quadrupole split components. The occurence of a pressure hysteresis is clearly observed.

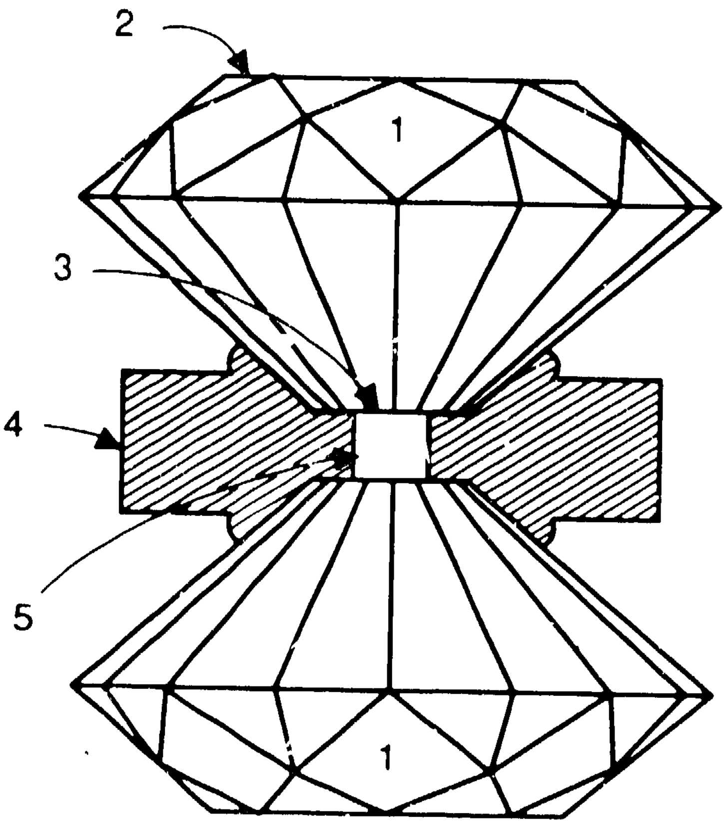


Fig. 1

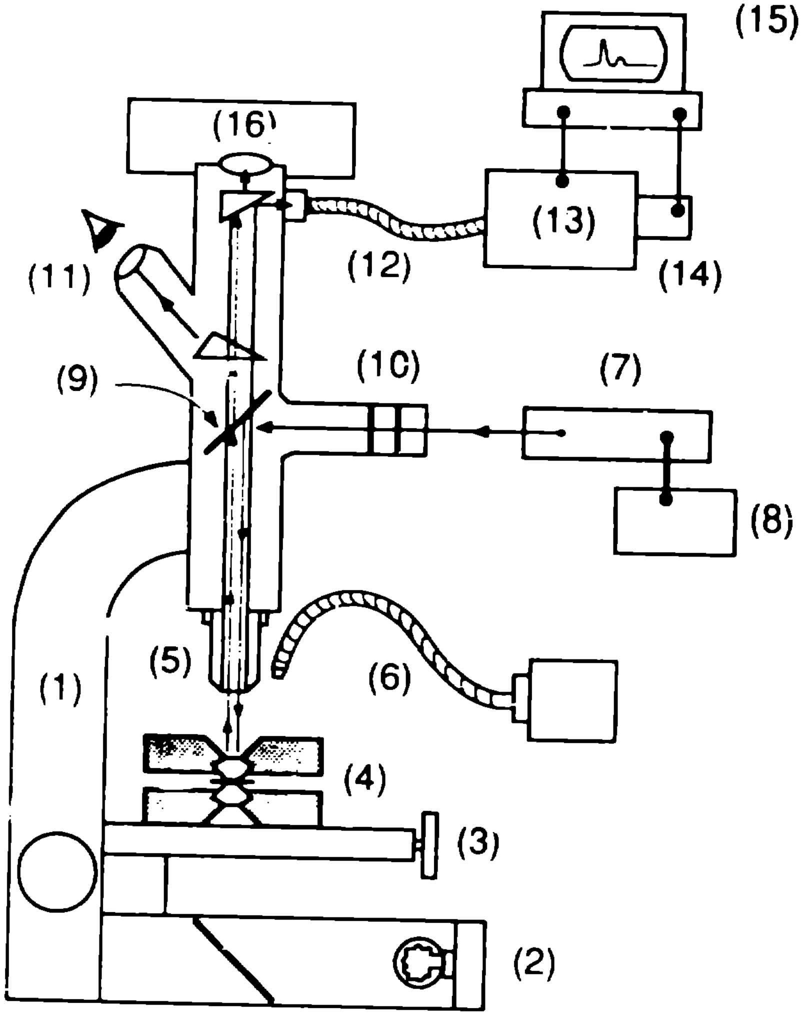


Fig. 2

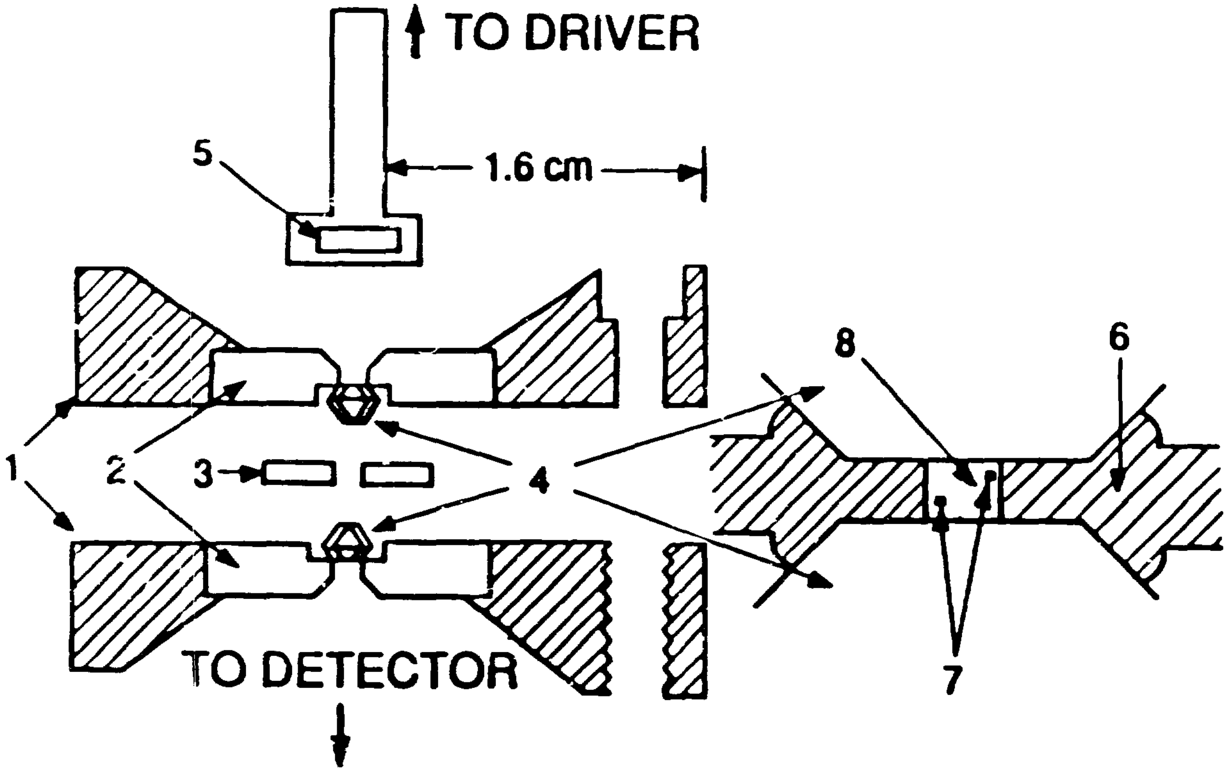
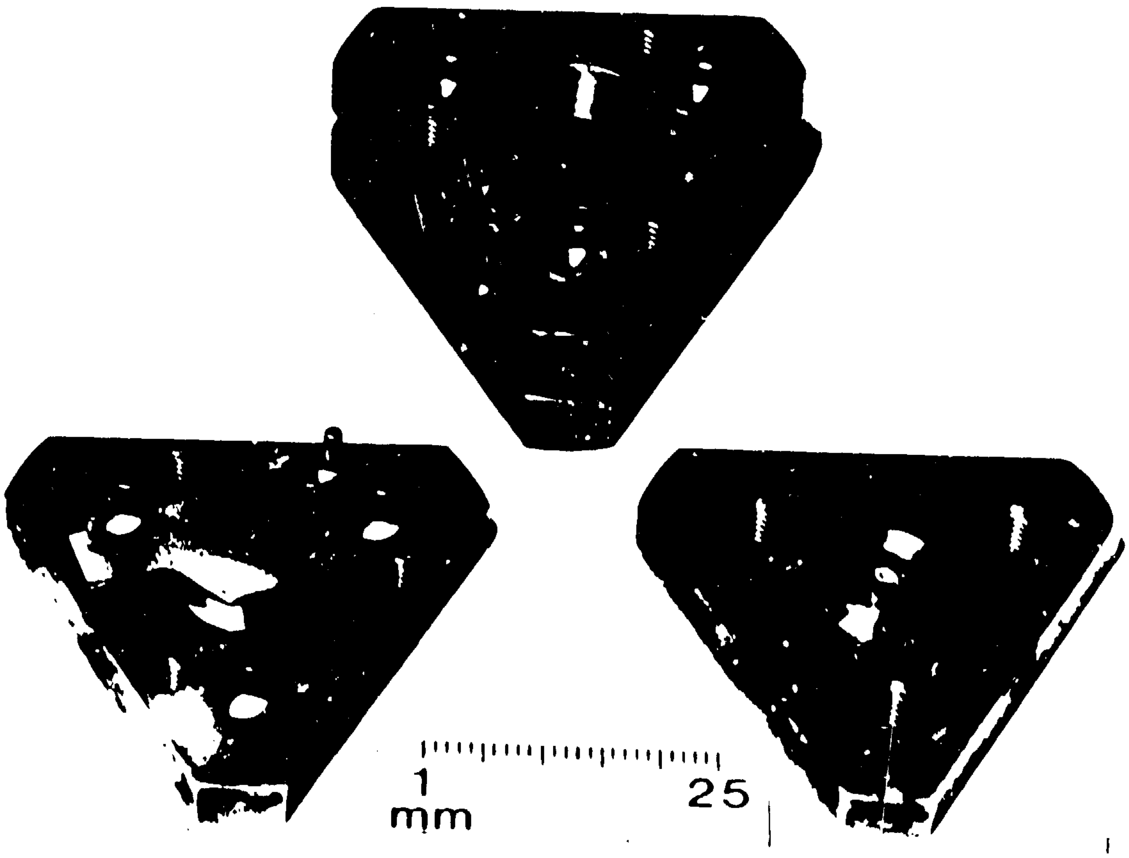


Fig. 3



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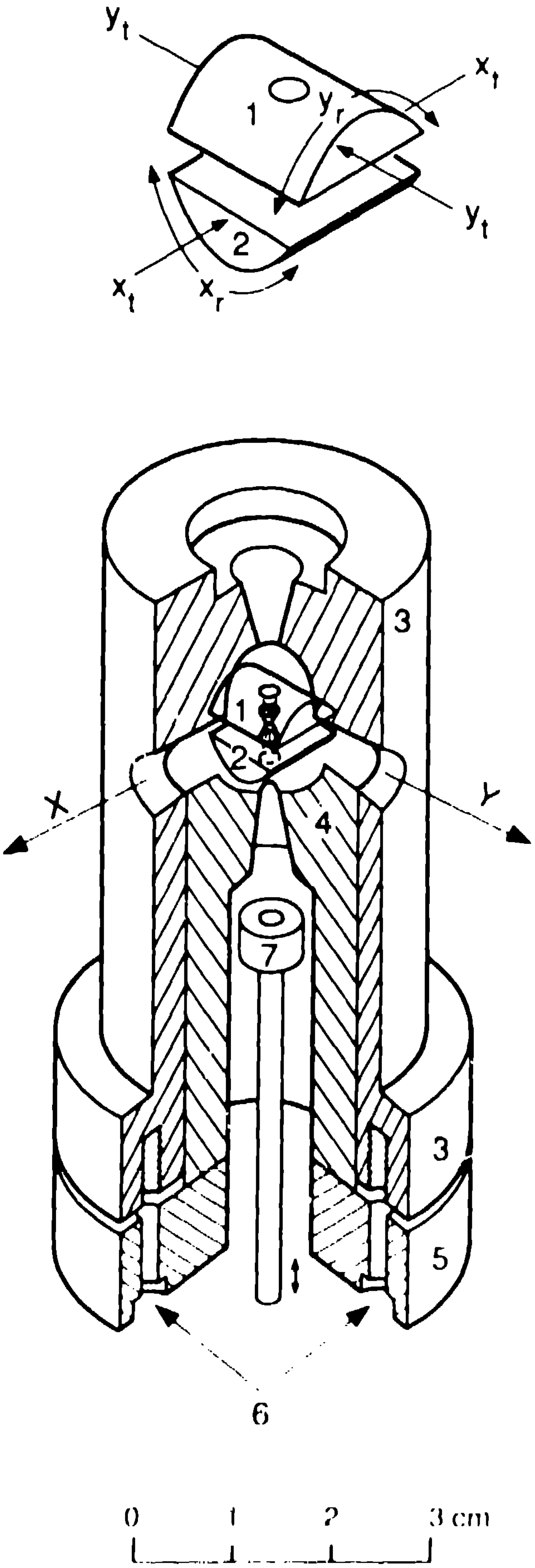

Fig.5

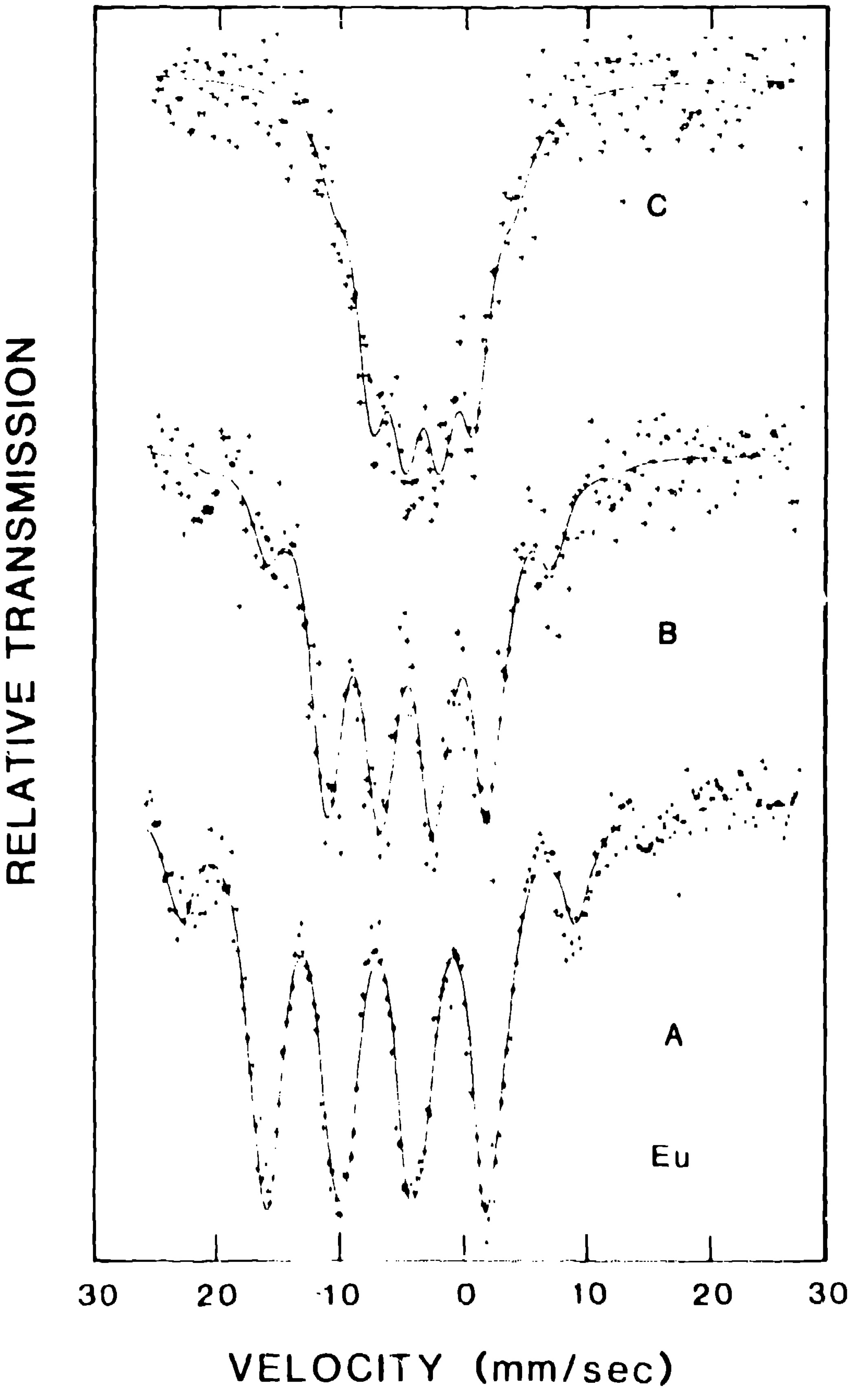
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Fig. 6

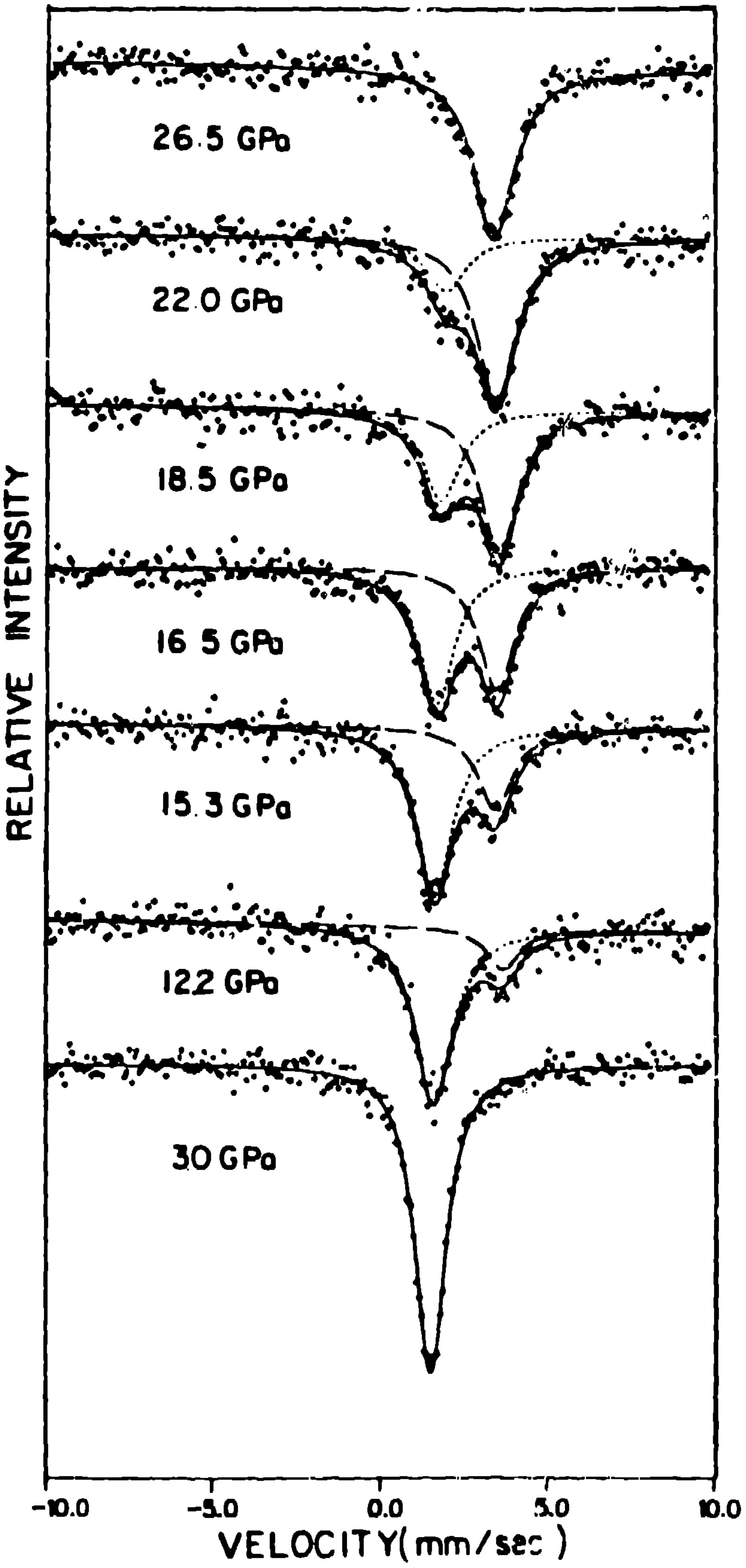
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Fig. 7

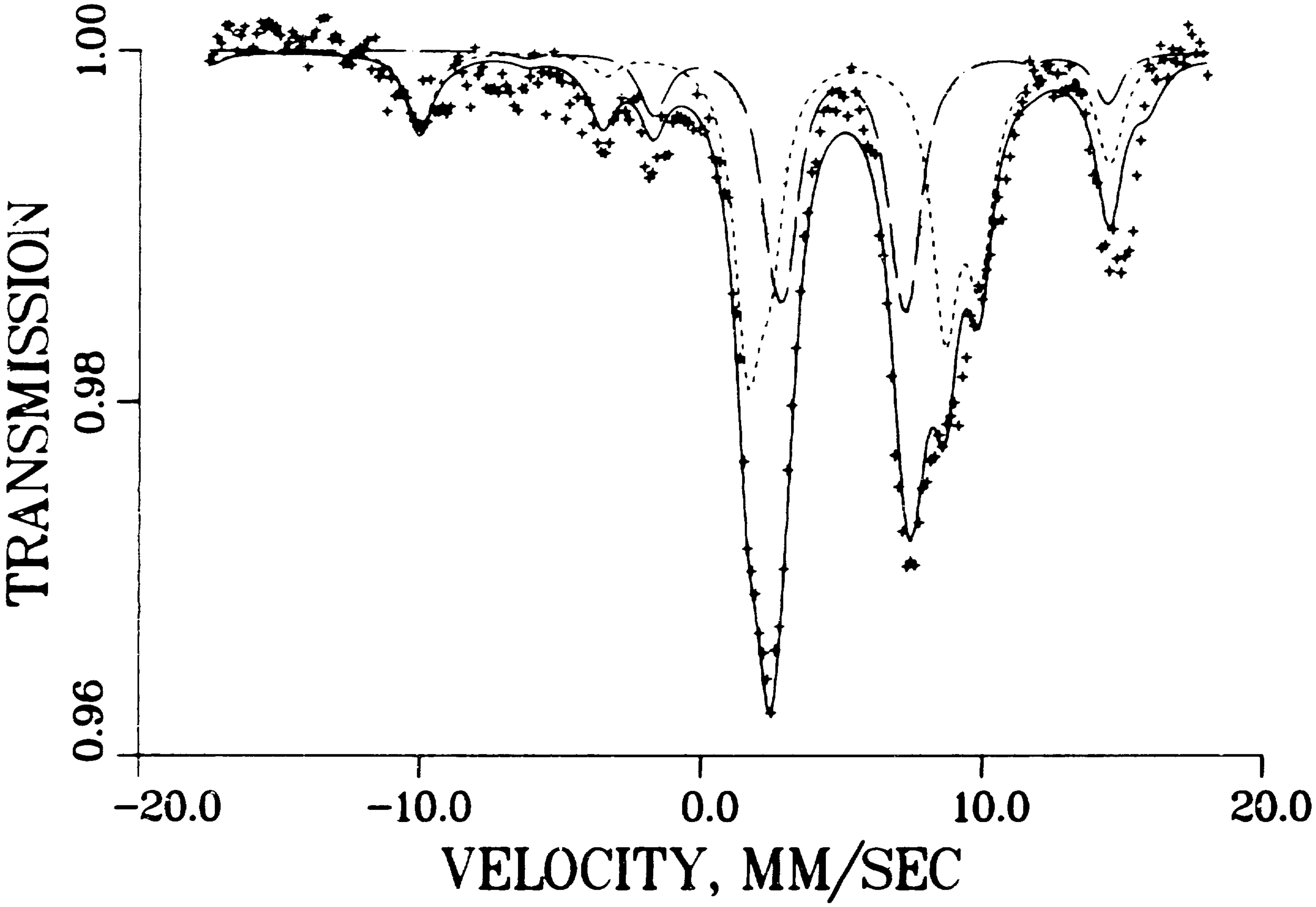
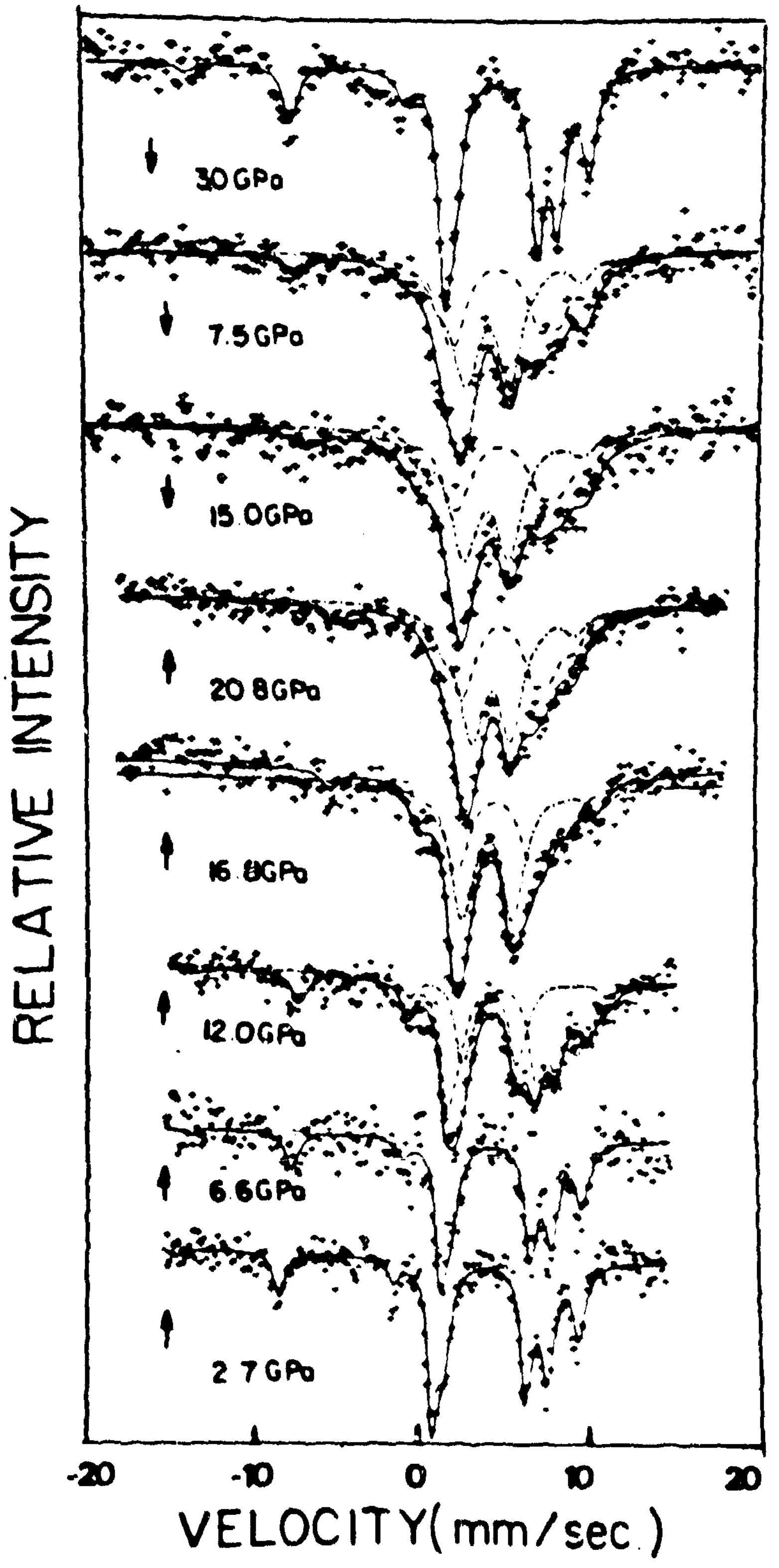


Fig.8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-10
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VELOCITY(mm/sec.)

Fig.9