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INELASTIC SCATTERING IN CONDENSED MATTER  
WITH HIGH INTENSITY MÖSSBAUER RADIATION

Progress Report  
for Period March 1, 1990 - September 30, 1990

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## ABSTRACT

We give a progress report for the work which has been carried out in the last three years with D.O.E. support. A facility for high-intensity Mössbauer scattering is now fully operational at the University of Missouri Research Reactor (MURR) as well as a facility at Purdue, using special isotopes produced at MURR. High precision, fundamental Mössbauer effect studies have been carried out using scattering to filter the unwanted radiation. These have led to a new Fourier transform method for describing Mössbauer effect (ME) lineshape and a direct method of fitting ME data to the convolution integral. These methods allow complete correction for source resonance self absorption (SRSA) and the accurate representation of interference effects that add an asymmetric component to the ME lines. We have begun applying these techniques to attenuated ME sources whose central peak has been attenuated by stationary resonant absorbers, to more precisely determine interference parameters and line-shape behavior in the resonance asymptotic region. This analysis is important to both the fundamental ME studies and to scattering studies for which a deconvolution is essential for extracting the correct recoilless fractions and interference parameters. A number of scattering studies have been successfully carried out including a study of the thermal diffuse scattering in Si, which led to an analysis of the resolution function for gamma-ray scattering. Also studied was the anharmonic motion in Na and the satellite reflection Debye-Waller factor in TaS<sub>2</sub>, which indicate phason rather than phonon behavior. We have begun quasielastic diffusion studies in viscous liquids and current results are summarized. These advances, coupled to our improvements in Microfoil Conversion Electron spectroscopy lay the foundation for the proposed research outlined in this request for a three-year renewal of D.O.E. support.

## Progress Report

### A. General review of progress during last 3 year period.

During the last grant period the successes of the research program can be summarized as follows:

1. Bruce Bullard has completed his thesis research and will receive his doctoral degree from Purdue University in December. A copy of his Ph.D. thesis is enclosed with this renewal proposal. Within the last year five publications related to his doctoral studies have been completed and two of these (submitted to Physical Review) are enclosed as preprints and are included as Appendices A and B of this renewal proposal. Also, Ammar Djedid completed his doctorate at Purdue in May of 1988.
2. Currently three graduate students are carrying out work related to this grant: Ralph Wagoner (Purdue), Scott Dickson (Purdue), and John Day (Purdue). Only one of these is currently being supported (Wagoner) and we seek support for all three, since the success of the proposed research depends on having adequate top-notch students. Some University of Missouri students have participated on the project, but we are presently seeking a well qualified U.M. student to pursue Ph.D. research in this area.
3. The last grant period has resulted in fourteen publications <sup>1-14</sup>. Two of these<sup>13,14</sup> have just been submitted and the manuscripts are reproduced as Appendices A and B.
4. This work has attracted wide spread attention from the scientific community, and several invited talks in Europe, New Zeland, Australia, and the United States have been given (See listing of invited talks at the end of this section).
5. European scientists have been particularly enthusiastic about this work and one of us (J.G.M.) has received recent letters from Prof. Dr. F. Wagner (Müchen, FRG) and Prof. K. Rubenbauer, Cracow, Poland, proposing collaborative research. Rubenbauer

is seeking a Fulbright fellowship to spend a year at Purdue working on this D.O.E. sponsored project.

6. Several European scientists have noted that the MURR facility is especially well suited to this work, and that there is not a facility of equal capability for this type of work in Europe. For example, the high flux reactor at Grenoble, using heavy-water, is only suitable for neutron beam experiments in scattering and cannot be used for fabricating high intensity ME sources of the type made at MURR.

#### List of Publications covering D.O.E. Grant Period 1988-1990

1. J.G. Mullen, A. Djedid, D. Cowan, G. Schupp, M.L. Crow, Y. Cao, and W.B. Yelon, *Physics Letters A* **127**, 242 (1988). "Representation of Lineshape Parameters and Deconvolution of Mössbauer Spectra."
2. J.G. Mullen, A. Djedid, B. Bullard, G. Schupp, D. Cowan, Y. Cao, M.L. Crow, and W. Yelon, *Hyperfine Interactions* **40**, 123 (1988). "Precise Determination of Mössbauer Lineshape Parameters Including Interference."
3. J.G. Mullen, A. Djedid, G. Schupp, D. Cowan, Y. Cao, M.L. Crow, and W.B. Yelon, *Physical Review* **B37**, 3226 (1988). "Fourier-Transform Method for Accurate Analysis of Mössbauer Spectra."
4. G. Haley, J.G. Mullen, and J.M. Honig, *Solid State Communications*, **69**, 285 (1989). "First Order Change in Hyperfine Interaction at the Verwey Transition in Magnetite."
5. M.L. Crow, G. Schupp, W.B. Yelon, J.G. Mullen, and A. Djedid, *Physical Review* **B39**, 909 (1989). "Temperature Dependence and Anharmonicity of the Debye-Waller Factor in Sodium Metal Using Mössbauer Gamma-ray Diffraction."
6. B.R. Bullard, J.G. Mullen, and G. Schupp, *Hyperfine Interactions* **55**, 1127 (1990). "Using Lineshape to Precisely Determine / recoil-free Fraction: Application to Tungsten. "
7. R. Wagoner, B. Bullard, M. May, S. Dickson, and J.G. Mullen, *Hyperfine Interactions* **58**, 2687 (1990). "Time Dependence of the Source Recoilless Fraction for a Cobalt-57 in Rhodium Source."
8. S.R. Hong, G. Haley, and J.G. Mullen, *Hyperfine Interactions* **57**, 2221 (1990). "Iron Mössbauer Spectra of Lava from Jeju Island and its Similarities to Moon Basalts."

9. B.R. Bullard and J.G. Mullen, Nucl. Instr. and Meth. B51, 198 (1990).
10. G. Schupp, B. Hammouda, and C.M. Hsueh, Phys. Rev. A41, 5610 (1990).
11. B. Hammouda, G. Schupp, and S. Maglic, Accepted for publication in J. Chem. Phys., "Quasielastic Gamma-Ray Scattering from Polydimethylsiloxane in Benzene Solutions."
12. J.G. Mullen, B.R. Bullard, and G. Schupp, Proceedings of the Zakopane School of Physics, accepted for publication as a part of the proceedings, World Scientific Publishing.
13. B.R. Bullard, J.G. Mullen, and G. Schupp, Submitted for publication in the Physical Review, "Mössbauer Line-Shape Parameter for  $^{183}\text{W}$  and  $^{191}\text{Ir}$  in Metallic Tungsten and Iridium.
14. B.R. Bullard and J.G. Mullen, Submitted for publication in the Physical Review, "Mössbauer Line-Shape Parameters for  $^{159}\text{Tb}$  in  $\text{TbAl}_2$  and  $\text{Tb}_4\text{O}_7$ .

#### Ph.D. Theses Resulting From The D.O.E. Sponsored Research

1. Bruce Bullard, "Mössbauer Studies in Tungsten, iridium, and Terbium Using Line-Shape Analysis." Purdue University, December 1990.
2. Ammar Djedid, "Precise Lineshape Measurements Using an Analytic Mössbauer Function." Purdue University, May 1988.
3. Lowell Crow, "Mössbauer  $\gamma$ -ray Scattering in Silicon and Sodium". University of Missouri, December 1987.

#### Invited Talks J. G. Mullen (1987 - Present)

1. Fourier Transform Mössbauer Spectroscopy: A New Tool for the study of Fundamental Physics, University of Texas at El Paso, May 26, 1988.
2. Testing Nuclear Dispersion Theory, University of Missouri at Columbia, March 7, 1988.
3. Precision Determination of Mössbauer Lineshape and Quantum Interference, Johns Hopkins University, March 2, 1988.
4. Gamma Ray Scattering, The University of Missouri University Research Reactor Facility, September 29, 1987.
5. True Mössbauer Lineshape and the Determination of Quantum Interference, Purdue University, September 3, 1987.

6. Precision Determination of Mössbauer Lineshape Parameters Including Quantum Interference, The University of Auckland, New Zealand, August 11, 1987.
7. Lineshape Determination in Mössbauer Spectroscopy, The University of Missouri Research Reactor Facility, June 6, 1987.
8. Mössbauer Diffraction Experiments at the Research Reactor Facility, University of Missouri at Rolla, Chemistry Department, May 2, 1987.
9. Precise Determination of Mössbauer Lineshape Parameters Including Interference, ICAME 87, Melbourne, Australia, August 16-21, 1987.
10. Unique Signatures in Mössbauer Spectroscopy and Deconvolution of Spectra, Jet Propulsion Lab, June 10, 1987.
11. Using the Fourier-Transform Method of ME Spectroscopy for Precision Determination of the Recoilless Fraction, Zakopane School of Physics, May 2, 1990.
12. Measuring Recoilless Fractions and Interference Parameters Using Super-Intense Sources of Tungsten - 183, Johannes - Gutenberg University, May 8, 1990.
13. Probing Fundamental Physics with the Mössbauer Effect, Purdue University, August 27, 1990.

#### Invited Talks G. Schupp (1987 - Present)

1. Dynamics of Polydimethylsiloxane in Benzene Solutions by Quasielastic Gamma-Ray Scattering, San Francisco, November 6, 1989.

## B. Line-Shape Studies

From the super-intense sources we have been able to fabricate at the MURR facility, we have measured Mössbauer spectra with an accuracy previously unattainable. By fitting our data to an analytic expansion of the convolution integral we have been able to determine the Debye-Waller factor (recoilless fraction) to unprecedented accuracy, accuracies comparable to the best specific heat data and having sufficient precision to complement specific heat data in testing and evaluating lattice dynamics calculations.

When source resonance self absorption (SRSA) is negligible there are six parameters that characterize the ME line shape. One of these is the interference parameter,  $\beta$ . By fitting our high precision data to the analytic expansion of the convolution integral, we have been able to make a quantitative test of the theory<sup>1,2</sup> of final state effects describing the asymmetric component of the ME spectral line. While our results agree with theory to about 10 to 20%, this difference is very significant in measurements of possible time reversal invariance violation for electromagnetic decay in nuclei, such as the extensive Cal Tech measurements,<sup>3</sup> where in the case of <sup>191</sup>Ir the contribution from final state effects is much larger than the sought time reversal phase parameter,  $\eta$ . For this case the value of  $\beta$  (E2) and  $\beta$  (M1) are so critical that the two theoretical calculations, which themselves agree to within 5%, that have been carried out lead to different conclusions as to whether or not there is a violation of time reversal symmetry for this case of mixed E2 and M1 decay.

We have carried out these studies of interference and recoilless fraction for <sup>183</sup>W in tungsten metal, <sup>191</sup>Ir in iridium metal, and <sup>159</sup>Tb in TbAl<sub>2</sub> and Tb<sub>4</sub>O<sub>7</sub>, fitting our data to the convolution integral, and making constrained fits to two more spectra simultaneously to reduce the effects of correlation between the line-shape parameters.<sup>4</sup>

In addition to precise values for interference and recoilless fraction, we find for one of the lineshape parameters, the intrinsic width of the ME transition, values accurate to about 1%, and we believe that our inferred lifetime for the isomer states are the most accurate to date.

These results are described in detail in Appendix A and B, and these two appendices



have been submitted as manuscripts for consideration as publications in the Physical Review B.

One of the important results to come from our studies of line-shape is a recognition of the importance of source resonance self absorption (SRSA) to the ME spectrum. We have derived an analytic expression<sup>5-9</sup> for the reduction in the emission recoilless fraction, and we find<sup>9</sup> that for the commonly used <sup>57</sup>Co in rhodium metal sources a surprisingly large time dependence to the recoilless fraction emitted from such a source, a point which has been ignored by people doing <sup>57</sup>Fe Mössbauer spectroscopy.

## References

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6. J.G. Mullen, A. Djedid, B. Bullard, G. Schupp, D. Cowan, Y. Cao, M.L. Crow, and W. Yelon, Hyperfine Interactions **40**, 123 (1988).
7. J.G. Mullen, A. Djedid, D. Cowan, G. Schupp, M.L. Crow, Y. Cao, and W.B. Yelon, Phys. Lett. **127**, 244 (1988).
8. J. G. Mullen, B. R. Bullard, and G. Schupp, Proceedings of the Zakopane School of Physics, Condensed Matter Studies by Nuclear Method, World Scientific Publishing.
9. R. Wagoner, B. Bullard, M. May, S. Dickson, and J.G. Mullen, Hyperfine Interactions, **58**, 2687 (1990).

### C. Attenuated Source Experiment

In an attempt to make better measurements of the interference parameter  $\beta$ , we have used a geometry used earlier by Mössbauer and co-workers.<sup>1</sup> An absorber of thickness  $t_1$ , is fixed in place relative to the source while an absorber  $t_2$  is Doppler shifted in the usual manner, so that the gamma beam arriving at the detector has passed through first the stationary and then the moving absorber. The first absorber primarily removes those gamma's centered about the transition energy  $E_o$ , and has less effect on those emitted in the wings, making the transmission signal more sensitive to asymmetries in the regions away from the center of the ME line, and giving a double peak for single line source and absorbers as shown in Figs. 1 and 2. Since the  $\beta$  parameter plays the dominant role in this asymmetry, the method allows a means of focusing on  $\beta$ .

The actual effect of a non-zero value of  $\beta$  can be seen directly in the data shown in Figs 1 and 2. The difference in height of the twin peaks is a result of  $\beta \neq 0$ . The existence of two peaks rather than the normal single peak is due to the attenuation of gammas centered about  $E_o$ . The transmission signal for such an arrangement of absorbers is given by

$$C(x) = C_o \left\{ 1 - f_{so} + \frac{2f_{so}}{\pi} \int dx' \frac{e^{-t_1 \mathcal{L}(2x)} e^{-t_2 \mathcal{L}(2x')}}{1 + 4X^2} \right\},$$

where

$$X = x' - x$$

and

$$\mathcal{L}(2x) = \frac{1 - 4\beta x}{1 + 4x^2},$$

where  $x$  is the reduced energy, i.e., in units of the level width  $\hbar/\tau$ .

To fit this function we have developed a method, described in the next section, that allows direct numerical fitting to any Mössbauer type convolution integral. We are presently testing this method using <sup>183</sup>Ta and <sup>183</sup>W metal absorbers. The data for four fixed enriched

$^{183}\text{W}$  and two moving natural  $^{183}\text{W}$  absorbers is given in Fig. 2. along with the best fit. As in the case of the analytic fit to single lines, good fits can be achieved with a single data set but the correlation between certain parameters leads to large errors in the value of the parameters. The solution to this correlation problem is to fit simultaneously more than one data set at a time.<sup>2</sup> We cannot only simultaneously fit two or more attenuated data sets, but can also simultaneously fit attenuated sets with data from standard moving and no fixed absorber type geometries. Collection of more  $^{183}\text{W}$  data is presently going on and results in this system are expected within the next three months.

## References

1. R.L. Mössbauer, H.E. Seelbach, B. Persson, M. Bent, G. Longworth, AEC Research and Development Report CALT-63-108.
2. B.R. Bullard, J.G. Mullen, and G. Schupp, *Hyperfine Interactions* **55**, 1127 (1990).

## Figure Captions

**Fig 1.** Inversion ME spectrum with two enriched stationary absorbers. The solid curve is the best fit to the data based on the convolution integral.

**Fig 2.** Inversion Me spectrum with four enriched stationary absorbers. The solid curve is the best fit to the data based on the convolution integral.

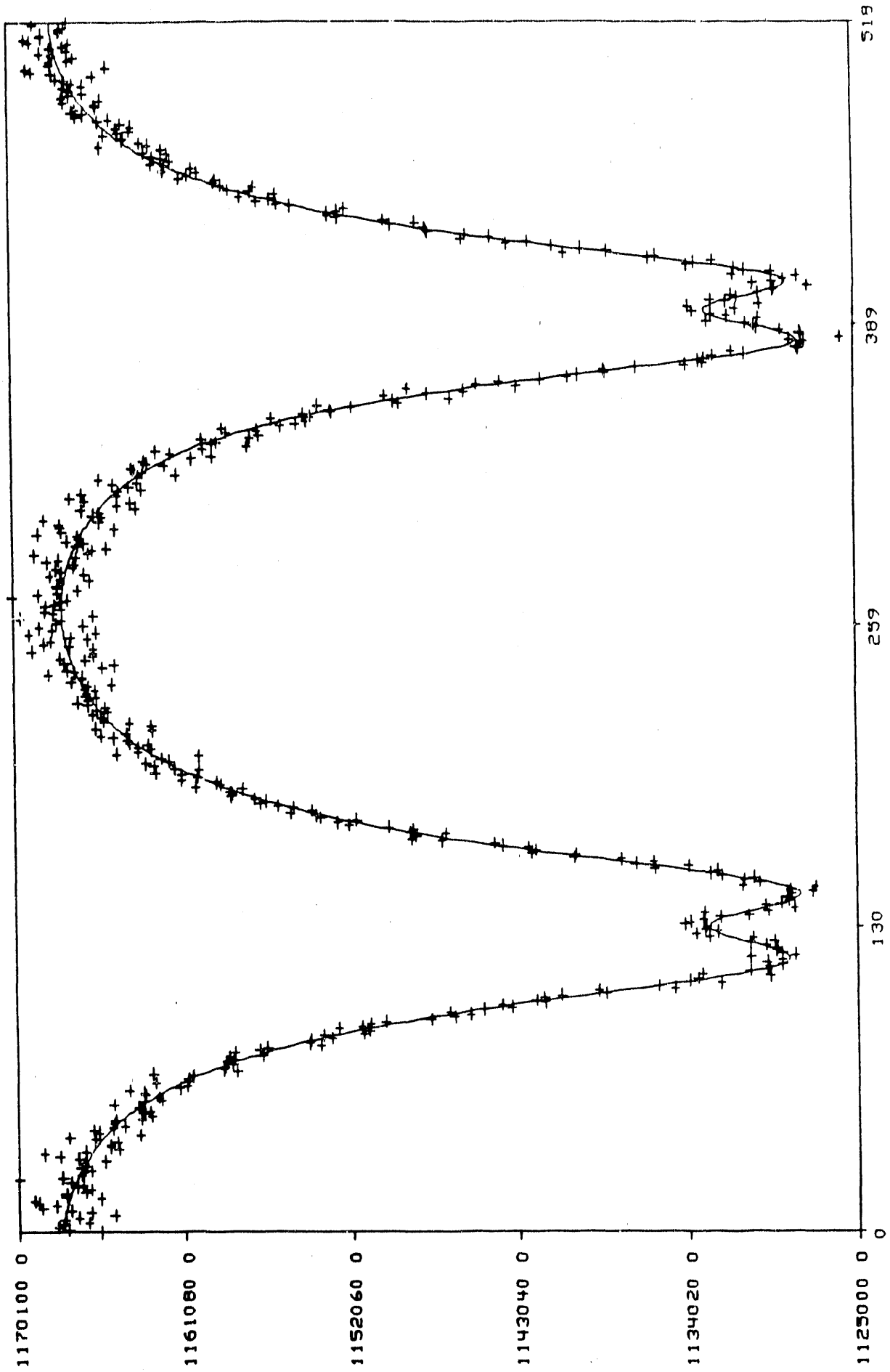


Fig. 1

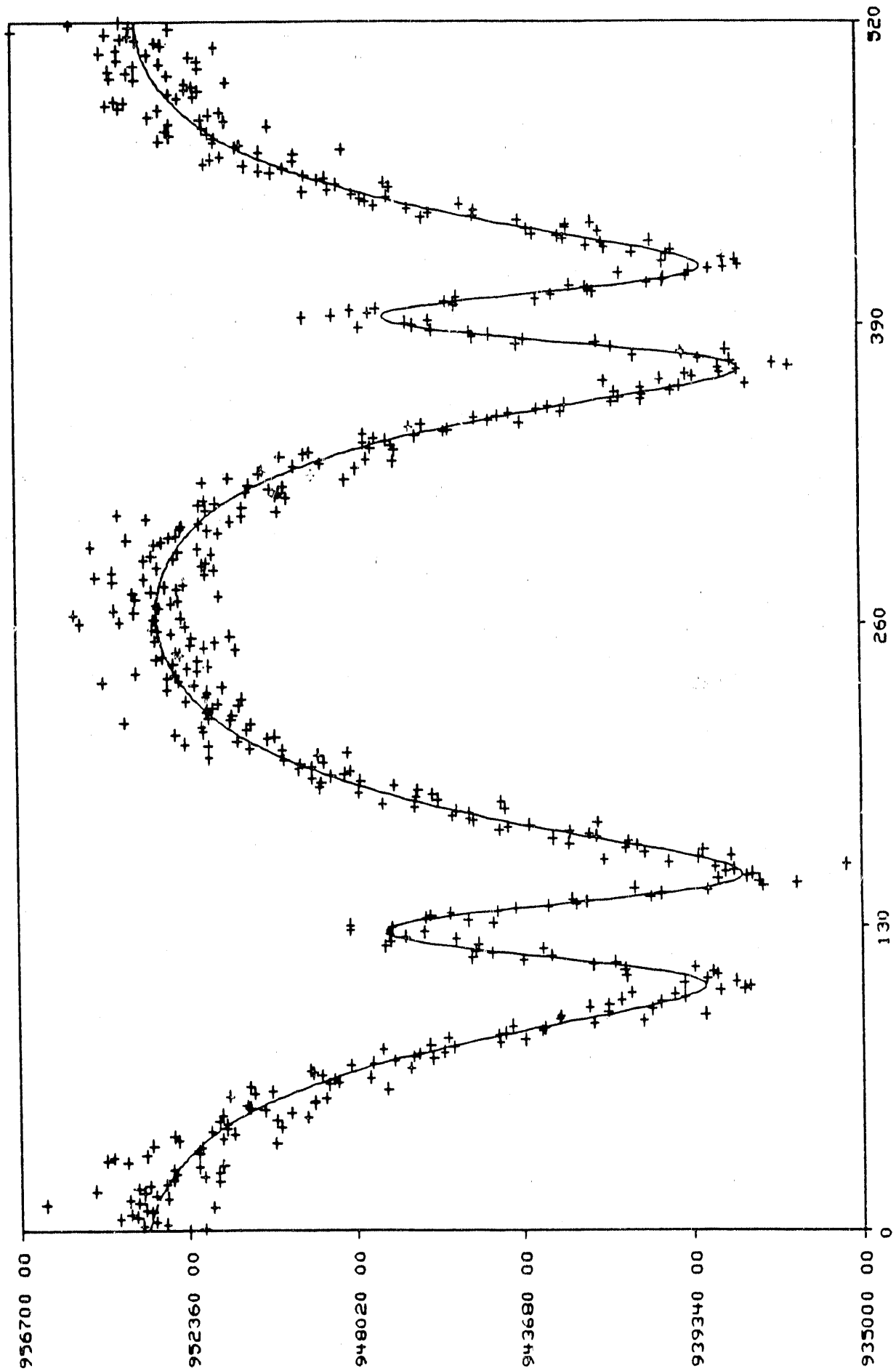


Fig. 2

#### D. A New Computational Method of Analysis and Its Applications

We have developed a method of numerical integration which uses a mix of both Weddle's rule and Gaussian quadrature using Legendre polynomials,<sup>1,2</sup> that allows us to accurately fit the data of any Mössbauer experiment. This includes standard cases where source and/or absorber have quadrupole or magnetic splitting as well as  $t_{rs}$ ,  $v_0$ , and  $\beta$ ; and also unusual cases of varying number, type, or geometry of absorbers or sources (the attenuated experiment mentioned before being an example).

As long as a mathematical expression can be written down for the transmission signal, and the integrand of the integrals in this expression approach zero as  $x$  goes to  $\infty$ , then our numerical method can be used to fit the data. There is no need to approximate the convolution integral, no need to restrict oneself to thin absorbers or widely shaped split lines, or replacing the absorber or source terms with expansions as a means of replacing the true convolution integral with some simpler version, although an analytic expansion of the convolution integral will reduce the computer time needed to achieve a good fit to data.

We have already tested the method using actual data from the attenuated source experiment, as well as  $^{183}\text{W}$  single line unbroadened data. The fits to the single line case were in excellent agreement with fits made using the analytic function developed earlier. We have also tested it against artificially generated data, including cases with large  $t_{rs}$ , and cases that included quadrupole splitting in the source, and in all cases accurate results were attained in reasonable computational times. It should be noted that in the case of a single unbroadened line, the analytical method is preferable due to its providing faster convergence. The time consumed, however, using the numerical method is by no means so long as to be impractical. For example, in the case of the attenuated source experiment with its complicated convolution integral, data for about 520 points, fit to 7 parameters, took about 4-5 hours of real time on our ISI/V24, and under 30 minutes on our Stardent Titan P2 mini-supercomputer. This technique is particularly useful in the majority of Mössbauer experiments presently being done which lack any exact analytical expression for their results.

Two applications of this method which we hope to utilize in the near future, are tests of the interference parameter used in time reversal experiments, and accurate measurement of parameters in various split spectra that are of current interest in Mössbauer studies.

Past time reversal experiments have all relied on theoretical calculations of an interference parameter  $\beta_T$ . This parameter is itself a sum of more basic terms,

$$\beta_T = \beta(M1) - \beta(E2),$$

the same two parameters on which the Mössbauer interference parameter  $\beta$  is dependent, i.e.,

$$\beta = \frac{\beta(M1) + \delta^2 \beta(E2)}{1 + \delta^2},$$

where  $\delta$  is the multiple mixing ratio<sup>3</sup>.

An accurate measurement of  $\beta$ , though not equivalent to measuring  $\beta_T$ , would none-the-less be a consistency check on the theoretical value. In fact, recent experiments<sup>4</sup> by our group suggest that the theoretical values for  $\beta$  in <sup>191</sup>Ir may be off by as much as 10%. This measurement was made using the standard Mössbauer geometry, and we hope the attenuated geometry will allow an even more accurate measurement of  $\beta$ , giving a more decisive check on the accuracy of present theoretical models. If the theoretical values are even slightly in error, then all future time reversal work would require either a new theoretical calculation of the value of  $\beta_T$ .

The great majority of present Mössbauer work involves quadrupole and magnetically split lines. Yet most of these studies still use approximations which have long been recognized as inaccurate. Measurements of  $\beta$  have particularly been neglected, and the assumption that  $n$  split lines can be fit to  $n$  Lorentzians, is an approximation that ignores the overlap between these lines as well as suffering from the inherent problems of fitting Lorentzians to lines that are not true Lorentzians to begin with. We plan to use our numerical method to draw out all the possible information available in Mössbauer studies, both the nuclear and solid state

parameters. We believe work in those cases, where the split lines are weakly resolved or not resolved at all, to be of particular importance since it is here that the approximations generally used in the field are most likely to give erroneous results.

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## E. Hyperfine Interactions Fitted to the Convolution Integral.

We have taken measurements of  $^{57}\text{Fe}$  in well characterized magnetite<sup>1</sup> as well as geological samples taken from Jeju Island.<sup>2</sup> These samples give interesting hyperfine split ME spectra. Although our first evaluation of these interesting spectra was carried out using the simple Lorentzian function commonly employed in obtaining hyperfine parameters, the residuals indicate that the fits to such data by this commonly used procedure is far from exact and we wish to now employ our direct fits to the convolution integral with a mini super-computer to reevaluate these data correcting for source resonance self absorption (SRSA) and eliminating the saturation effects associated with the use of absorbers of finite thickness.

Two interesting results which have already come from these studies is that the ME spectra for basalts from Jeju Island are remarkably similar to those found in lunar basalts. Secondly, we find contrary to all earlier studies that stoichiometric and homogeneous magnetite has a sharp change in the hyperfine field at the Verwey transition characteristic of a first order transition. This result is nicely collaborated by the extensive studies, including specific heat measurements carried out by J. Honig's group.

In addition to attempting to directly fit these hyperfine split spectra to the convolution integral using our new curve fitting procedures, we are also planning the systematic



deconvolution of these spectra using the Fourier transform technique described in a recent publication.<sup>3</sup> If this latter technique is successful it will make possible more accurate measurements of hyperfine spectra for all Mössbauer studies of internal magnetic fields and electric field gradients.

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## F. Burnout Cross Section of $^{159}\text{Dy}$

In our attempts to fabricate superintense Mössbauer sources, we have succeeded in several cases. An exception has been the interesting isotope  $^{159}\text{Dy}$ , whose daughter,  $^{159}\text{Tb}$  has an exceptionally broad width and is well suited to the study of low energy excitations. This surprising result was traced to the large burnout cross section for  $^{159}\text{Dy}$  which we have determined<sup>1</sup> to be  $8 \pm 2$  kilobarns. This high cross section impedes the production of the Mössbauer parent  $^{159}\text{Dy}$  by two orders of magnitude. Using the special facility at Purdue University we have measured the line-shape parameter of this isotope, despite its low intensity. This is possible because this transition is exceptionally clean and experiments can be done directly in the photon beam without the aid of a monochromating crystal filter.

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## G. Anharmonicity in the Temperature Dependence of the Debye-Waller Factor in Sodium Metal.

The Debye-Waller factor of sodium has been measured<sup>1</sup> as a function of temperature from 80 to 295 K using Mössbauer  $\gamma$ -ray scattering. The high energy resolution provided by this technique allowed experimental separation of the elastic scattering from the inelastic thermal diffuse scattering. The results were compared with the harmonic model using integrations over dispersion curves from the neutron-scattering measurements of Woods *et al.*<sup>2</sup> and the lattice-dynamics calculations of Glyde and Taylor.<sup>3</sup> The Debye-Waller exponent was shown to exceed the harmonic prediction by 23% at room temperature, and this difference is attributed to anharmonic terms in the interatomic potential.

### References

1. M.L. Crow, G. Schupp, W.B. Yelon, J.G. Mullen, and A. Djedid, *Phys. Rev.* **B39**, 909 (1989).
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## H. Recent Scattering Experiments

The efforts of the last year have been focused in two areas: quasielastic scattering from systems undergoing diffusive neutrons, and scattering close to the transition in a martensitic phase (NiAl). These have provided interesting new results summarized below, and have also made clearer the need for some major improvements in the experimental equipment in order that these studies be performed more reliably and efficiently. These will be explained in the proposal section.

Our Mössbauer diffraction instrument at MURR, with its intense sources, has allowed us to expand into the area of quasielastic gamma-ray scattering (QEGS). The  $\mu\text{eV}$  energy resolution, as well as the wide range of momentum transfer of this technique, opens new

possibilities for the study of molecular liquids in general and macromolecular liquids in particular, as well as for solid state diffusion.

Liquids are dominated by thermal Brownian type motion which leads to quasielastic (centered at  $\Delta E = 0$ ) scattering processes. Mössbauer studies of diffusion in liquids were first conducted for  $^{57}\text{Fe}$  ions dispersed in glycerol by Bunbury *et al.*<sup>1</sup> and by Craig and Suttin.<sup>2</sup> Subsequent to these measurements, Abras and Mullen,<sup>3</sup> Singh and Mullen<sup>4</sup> and, most recently, Nienhaus *et al.*,<sup>5</sup> have also studied different aspects of the scattering by glycerol. Until our work with QEGS, however, these studies have been limited to liquids which included the Mössbauer nuclide. In these studies, the resonant nuclei are moving with the diffusing medium and absorb the incoming radiation according to the velocity of the nuclei. In the present studies, the incoming radiation is Raleigh scattered by the diffusing medium itself and the data interpretation should be unambiguous. Figure 1 summarizes the main techniques used to investigate the dynamics of liquids<sup>6,7,8</sup> at various characteristic length scales going from the collision dominated hydrodynamic region (very small  $Q$  and  $\omega$ ) to the free streaming ideal gas limit (very large  $Q$  and  $\omega$ ). Here,  $Q$  and  $\omega$  represent the momentum and energy transferred during scattering. Typical characteristic lengths (Angstroms) and time scales (picoseconds) for liquids fall within the  $(Q, \omega)$  windows for neutron scattering, gamma-ray scattering and computer simulation (molecular and Brownian dynamics).

We have conducted three studies on viscous liquids using this new technique. The first was on pentadecane<sup>9</sup> (published in the Physical Review A, May 1990) and the second was on polydimethylsiloxane (accepted in July 1990 for publication in the Journal of Chemical Physics). In both of these studies, the quasielastic broadening of the Mössbauer velocity spectrum was observed by coherent scattering at the first liquid structure peak. For pentadecane, this broadening was observed versus temperature of the sample in the 12°C to 74°C range and in the polydimethylsiloxane case, it was observed versus dilution in benzene at room temperature. Also for pentadecane, the width of the quasielastic scattering was measured versus the scattering vector  $Q$  (by setting at different positions across the liquid

structure peak) at room temperature. This latter result showed deGennes narrowing<sup>10</sup> of the width when setting at the  $Q$  corresponding to the center of the liquid structure peak.

In the polydimethylsiloxane investigation, part of the scattering was found to be "elastic" (i.e., within the resolution of QEGS) and part quasielastically broadened. Upon dilution in benzene, the "elastic" fraction of the scattering decreased quickly, demonstrating a softening of the stiff degrees of freedom along the backbone of the polymer chain while the scattering from the softer modes broadened further.

The most recent quasielastic scattering measurements by us have been on glycerol in the temperature range from 24°C to 113°C. It is presently in the interpretation and manuscript preparation stage. Based on the earlier work noted on glycerol and the fact that it is a smaller, nearly spherical molecule with a small electric dipole moment, we expected that our results would be straightforward to evaluate. We found, however, a much greater broadening with temperature than was expected (see Fig. 2) and a more complete model is being sought.

\* \* \*

The martensitic transition in NiAl is seen as representative of a large class of materials such SrTiO<sub>3</sub>, KMnF<sub>3</sub> in which a soft phonon is observed above  $T_c$ . Below  $T_c$  a distorted structure is observed with the new Bragg peaks associated with the soft phonon position. In these systems, a "central peak" is seen at  $E = 0$  at the phonon wavevector and there has been much discussion and investigation of the nature of this central peak. QEGS is one of the best techniques for such an investigation since it has simultaneously excellent energy resolution and access to a large region of reciprocal space.

In NiAl, a soft phonon and central peak is seen at a  $\vec{q} \simeq \frac{1}{6} - \frac{1}{60}$  (and equivalent positions although the transformed structure appears to correspond to a  $q$  vector closer to  $\frac{1}{7}$ ). Our studies show a more complicated situation in which, rather than satellites along the line between the 2 2 0 and 3 1 0 with spacing  $\sim \frac{1}{7} - \frac{1}{70}$ , there are two lines of spots parallel to

this line, one closer to the origin, the other further away than the 2 2 0 - 3 1 0 line. This structural problem warrants further detailed study.

QEGS measurements were made at the strongest of these satellites as a function of temperature. Up to  $\sim 30$  K above  $T_c$ , the scattering was fully elastic. Above this point, the intensity was too low to follow further. This observation is consistent with other studies which fail to observe broadening or inelasticity in the central peak and suggests that the central peak may correspond to static domains, perhaps induced by strains well above  $T_c$ .

Both the liquid scattering studies and these emphasize the potential of the QEGS technique but both were hampered by limited intensity and less than optimum sources (especially in source cooling). We outline a number of improvements in the next section which will improve these studies substantially.

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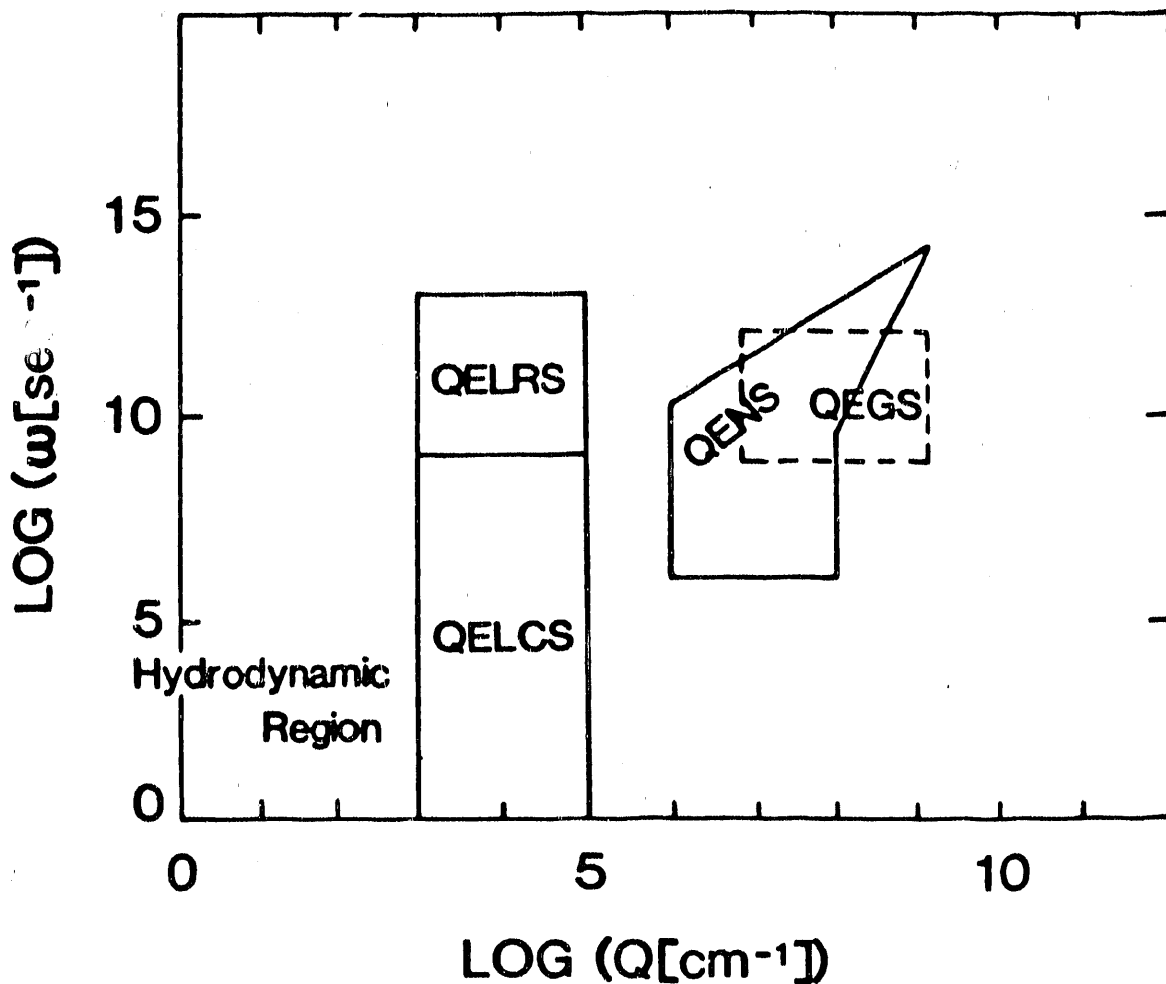


Fig. 1. Diagnostic methods used to investigate the dynamics of liquids: quasielastic light Rayleigh scattering (QELRS), quasielastic light correlation spectroscopy (QELCS), quasielastic neutron scattering (QENS), and quasielastic gamma-ray scattering (QEGS). The computer simulation region (molecular or Brownian dynamics) overlaps with QENS and QEGS.

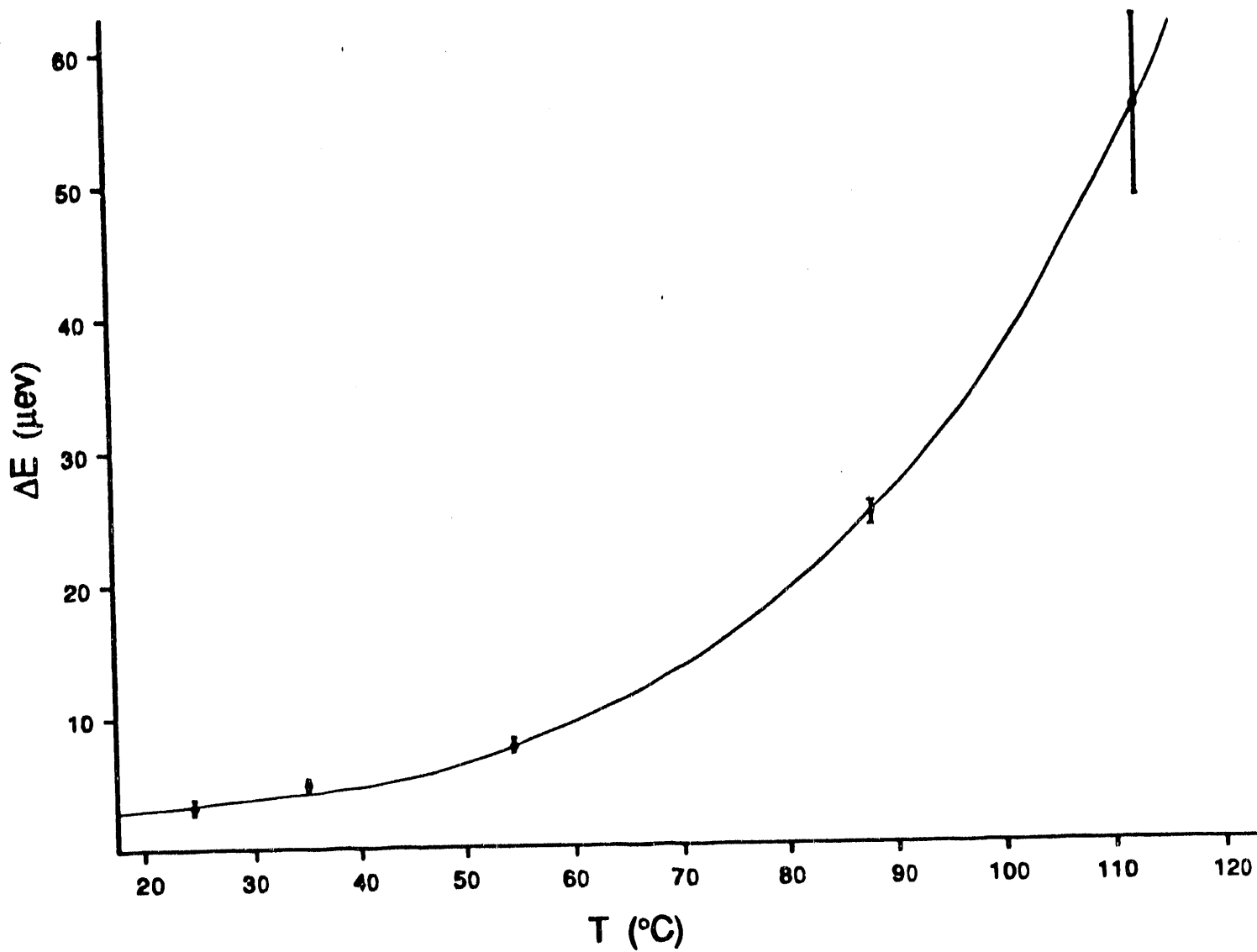


Fig. 2.  $\Delta E$ , the measured width of the quasielastically broadened line minus the elastic width, in  $\mu\text{eV}$  versus the temperature in  $^{\circ}\text{C}$ .

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