

## PROGRESS REPORT

## QUASIELASTIC SCATTERING FROM LIQUIDS:

Although quasielastic scattering from liquids by neutrons has been used as a powerful tool for investigating diffusion[33], we have been the first to demonstrate that Mössbauer photons can be used to study diffusion in the liquid state[15,16,29] (also see Appendix B).

We have carried out these first quasielastic gamma-ray scattering studies using intense ME sources of  $^{183}\text{Ta}$ . The first two investigations were done with pentadecane[15] and polydimethylsiloxane in benzene solutions[16]. In these measurements an elastic and quasielastic line is observed and the broadened line has been ascribed to diffusion. More recently we have studied diffusion in glycerol[29] (Appendix B), a somewhat simpler system than used in our first studies, and we found that the broadening observed was proportional to the temperature divided by the viscosity, which by the Einstein-Stokes relation is proportional to the diffusivity.

In contrast to the earlier Mössbauer studies of diffusion in liquids[34-38], these studies do not contain a ME ion in the solution. When scattering from such a pure liquid, the pair correlation function is crucial to the analysis instead of the self correlation function, and hence, the theoretical analysis of these quasielastic measurements is more difficult than the earlier ME studies of liquid diffusion, where the Singwi-Sjölander[40] theory is worked out in detail for the incoherent, self-correlation case.

## LATTICE DYNAMICS:

We have measured the recoilless fraction and Debye-Waller factors and have expanded on the earlier work of Heberle[40], showing the convergence of the Debye integral in both exponential and polynomial expansions. Below we give a broad review of these contributions. The reader is referred to the Appendices C,D, and F and References 17,19, and 26-28 for a more detailed review of our contributions in this area.

## RECOILLESS FRACTIONS:

Using a precision line-shape measurement, fitting the data directly to the transmission integral, we[17,19] measured the temperature dependence of the recoilless fraction from 80 to 1067K. These are to our knowledge the most precise recoilless fraction measurements ever made with an error of only about 1% at each temperature. The data fit a Debye model with a Grüneisen correction to account for thermal expansion effects. Only at the very highest temperature was a significant deviation

from experiment seen, and this could be attributed to quartic anharmonic effects.

Recoilless fractions, which are closely related to Debye-Waller factors when the ME atom is the same chemically as the host lattice, are quite distinct when the ME atom is an impurity in a host lattice. Recoilless fractions can serve a special role in the lattice dynamics of crystals containing impurities, where the local binding of the impurity to the lattice is sought. In such cases, the contributions to the Debye-Waller elastic scattering or the heat capacity may be negligible, but the contributions to the recoilless fraction can be quite large. An important example of impurity recoilless fractions is given in the section on DILUTE METAL ALLOYS discussed below.

#### DEBYE-WALLER FACTORS:

The ability to measure elastic scattering intensity, or Debye-Waller factors, to high precision gives another useful tool for studying lattice dynamics. In the past heat capacity data has played a much more significant role in evaluating lattice dynamical models than measurements of elastic x-ray scattering. This is no doubt due to the fact that heat capacity measurements have been much more precise than the measurements of the Debye-Waller factor.

As these two types of measurements are complementary, representing different moments of the phonon density of states function, both are important, distinct handles on model calculations of lattice dynamics. Supersources, coupled with line-shape analysis using the transmission integral, will allow measurements of the Debye-Waller factor to accuracies an order of magnitude better than in the past, and afford another useful tool for the study of lattice dynamics.

Using  $^{183}\text{Ta}$  we have measured the Debye-Waller factors sources for metallic sodium[10] and metallic silver[27] (see also Appendix C), and we are currently carrying out such measurements on the engineered material GaAs/AlAs[28] (see also Appendix D). In the first two cases we were able to make precision determination of anharmonic effects, while in the latter case we found a very large temperature dependence to the elastic scattering, which cannot be understood in terms of conventional theories.

One recent contribution to the determination of Debye integrals has been submitted to Hyperfine Interactions [26] (see also Appendix F). In this paper the work of Heberle[40] is expanded to show that it is much simpler to evaluate the direct exponential expansion of the Debye-type integrals using current computing capabilities, than to use multiple expansions as first proposed by Heberle. We evaluate the convergence of these Debye integral expansions as a function of the number of terms used and the time required. We show that the simple direct exponential expansion is simplest and best and gives results accurate to 0.01% in times of order or less than one second even using modest personal computers.

## DILUTE METAL ALLOYS:

In 1989 a startling paper was published by H. Shecter et al.[41], showing an anomalous break in the Debye-Waller factor for dilute  $^{119}\text{Sn}$  dispersed in metallic lead at temperatures about equal to one fourth of the melting temperature of pure metallic lead. A very recent article[42] restates the earlier results, and the effect is attributed to a local melting phenomenon about the tin impurity.

Because of the studies that we had made on recoilless fractions, we remeasured the ME for dilute tin in lead in an attempt to confirm the earlier reported results[41,42]. The first report on our results has been submitted to ICAME '93 and is given in detail in Appendix E. In contrast to Shecter et al. we find no anomalous cut-off in the Debye-Waller factor over the entire temperature region from 4-300K. Our very dilute samples with 1.3, 1.6, and 3.1 at.% all gave a Debye-Waller temperature dependence with a characteristic temperature of 117K, while our high concentration sample gave a significantly higher characteristic temperature of 131K. The latter case we attributed to a precipitation phenomenon with small precipitates of Sn in the lead host matrix causing the enhanced binding.

In none of our data is there any evidence of a sudden, precipitous fall off from the Debye-Waller behavior as reported by SSYBZ[41,42]. Our measurements (see Appendix E for detailed graphs displaying our results) indicate that samples containing 3 at. % tin or less are dispersed and do not show a sudden fall-off in  $f(T)$  as the temperature rises. Our 5.7 at. % sample has a distinctly lower slope to the  $\ln f$  vs.  $T$  curve. At temperatures above room temperature a drop in  $f(T)$  may occur as the tin dissolves into the lead host. The sample preparation technique used in this research is quite different from that of SSYBZ. Perhaps this difference is the source of the discrepancy between the two results. In these studies we spent much time confirming the composition and the homogeneity of our samples, using a tunneling microscopy and wet chemistry techniques.

In our Mössbauer spectra (see Appendix E) there exists a large enhancement in the line width for our 1.6 at. % sample, a broadening on the order of 50% at 4.5K which persists in some data runs up to about 50K. This sample had been measured after cold rolling without anneal.

We do not have enough data on this low temperature line broadening at this time to warrant lengthy speculations, but a recent proposal of spin density waves (SDW) in lead by Overhauser[43] could serve as a possible explanation of the effect. Overhauser postulates a SDW with nodes at the lattice sites. Thus, an annealed sample would not show any hyperfine interactions, while a severely strained sample could show an effect resulting from the tin ion being slightly displaced from the equilibrium lattice of the host material. At higher temperatures, then, the line will narrow due to thermal motion.

As of this date, several experimental runs have been completed on both annealed and unannealed samples. Our results are consistent with the Debye model using a

simple impurity mass correction. The effective Debye temperature for dispersed tin in lead is found to be 117(2)K over the temperature interval 4K to 295K. In contrast to the SSYBZ report, we find no anomalous fall-off from Debye-Waller behavior. Higher concentrations of Sn above the solubility limit may well lead to sharp bends in the  $f(T)$  curve, which would be interesting as a tool for the study of precipitation, but does not support the original premelting model advanced by SSYBZ. Future work will seek to clarify the source of this discrepancy as well as investigate the possible effects of spin-density waves.

One exciting application that this study could result in is a better understanding of low temperature precipitation phenomena. Composite materials are very much at the forefront of materials technology, and ME spectroscopy would be a powerful new tool for studying low temperature precipitation, where the kinetics are so slow that macroscopic approaches are ineffective.

### **METHODOLOGY:**

In developing the capability of handling and utilizing the most intense Mössbauer sources ever fabricated, we have been able to also develop some firsts in ME methodology. We are the first to use Soller Collimators in ME spectroscopy, the first to use monochromating filters to filter out unwanted radiations from our beam, and the first to use moving monochromating filters as a means of Doppler shifting the Mössbauer line with both the source and resonant detector stationary. Also we have demonstrated that it is possible to tune the isomer shift to any sought value by moving the monochromating filter and simultaneously looking at the ME spectrum with a moving absorber. These advances in methodology were given by the Principal Investigator as an invited talk at the XXVIII Zakopane School of Physics. The paper on this work is reproduced in Appendix A, and gives a nontechnical summary of these advances. This paper has been accepted for publication and will appear as a part of the Proceedings of this School.

### **LINE SHAPE AND ITS ROLE IN FINDING MATERIALS AND NUCLEAR PARAMETERS:**

The success of this entire program has depended critically on the development of a detailed understanding of the role of ME line shape on basic physical quantities like the recoilless fraction and line position. By this approach we have been able to determine recoilless fractions and Debye-Waller factors to accuracies one order of magnitude better than earlier work.

As a spin-off of this work we have also found accurate values of interference parameters, internal conversion coefficients, and nuclear lifetimes. Our most recent publications dealing with this subject are given in references 17-20 and 23. Although

there are many exciting things that we could still profitably do related to basic issues in ME line shape, we have basically completed those aspects of this subject that are essential to the completion of the mission of the grant.

We should mention that one of the most significant results of our interference parameter measurements is the discovery that the Mössbauer interference parameter differs from values predicted by theory[44,45] by approximately 10%. This has deep implications for those who wish to do time reversal invariance experiments, such as the one carried out by Gimlett, et al. [46].

The methodology of fitting ME spectra to the convolution integral will still play an important role in our future work, but it will be as a tool to probe materials physics, and new results related to nuclear and fundamental physics will be incidental.

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