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SEARCH FOR THE KOHN EFFECT IN LIQUID LEAD

P. D. Randolph

IDAHO NUCLEAR CORPORATION
NATIONAL REACTOR TESTING STATION
IDAHO FALLS, IDAHO

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SEARCH FOR THE KOHN EFFECT IN LIQUID LEAD

ABSTRACT

Recent theoretical calculations have shown that the so-called Kohn anomaly may be observable by neutron scattering in liquid metals, thus furnishing a new way of measuring the Fermi Surface in these materials. The effect is expected to appear as a discontinuity in a curve of quasi-elastic widths as a function of neutron momentum transfer. Using the MTR phased chopper velocity selector we have made slow neutron scattering measurements of the quasi-elastic widths in liquid lead at 350°C. These measurements were made at six incident energies in the range $.015 \leq E_0 \leq .025$ eV and span momentum changes $h\kappa$ in the range $1.8 \leq \kappa \leq 4.5 \text{ \AA}^{-1}$. To within the accuracy of the experiment no Kohn anomaly was observed. The variation of the widths agrees with the calculations of DeGennes at the first diffraction maximum and minimum, but not at large κ .

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Search for the Kohn Effect in Liquid Lead

I. INTRODUCTION

It was first pointed out by Kohn⁽¹⁾ several years ago that the presence of a logarithmic singularity in the static dielectric function of a metal crystal should give rise to a small discontinuity in the slope of the phonon dispersion curve $\omega(\vec{q})$ of the crystal. Here ω is the phonon frequency and $\vec{q}$ its wave vector. This discontinuity, the so-called Kohn effect or anomaly, would appear at a phonon wave vector $\vec{q}$ of

$$|2\pi\vec{\tau} + \vec{q}| = 2k_F$$

where k_F is the magnitude of the wave vector for electrons at the Fermi Surface of the crystal and τ is a vector of the reciprocal lattice. The predicted effect was first observed experimentally by Brockhouse et al⁽²⁾ in crystalline lead, and has since been observed in other metals⁽³⁾. The effect occurs whenever the phonon wave vector is equal to an extremal distance across the Fermi Surface. In principle then, by measuring the dispersion curves in many directions in the crystal and observing the Kohn anomalies, one could map the Fermi Surface for the material. Since the Fermi Surface in a solid has in general a quite complex shape, this would be a very lengthy process. The usual way of obtaining the Fermi Surface in crystals is by the de Haas-van Alphen effect or by ultrasonic attenuation experiments. Both of these later methods must be done at liquid helium temperature, and therefore cannot be used for liquid metals.

In the case of liquids the method of positron annihilation at present offers the only method of measuring the Fermi radius⁽⁴⁾.

It has been suggested recently by Singwi and Andersson⁽⁵⁾ that the Kohn effect might also be observable in liquid metals by neutron scattering. However for the liquid case, the effect would be observed in the widths of the quasi-elastic peaks of the scattered neutron spectra rather than in the dispersion curves. According to Singwi the quasi-elastic peak width ΔE , when considered as a function of the neutron momentum transfer $\hbar\kappa$, should have a discontinuity in its slope at $\kappa = 2k_F$. The effect is expected to be rather small in any case, and if the Fermi Surface is not fairly sharply defined, it may not be observable at all. Generally speaking, if the free electron for a liquid metal is valid, then the Fermi Surface is expected to be sharp, and the value of k_F is given by

$$k_F = (3\pi^2 n)^{1/3}$$

where n is the number of free electrons per unit volume of the liquid. An observation of the Kohn effect that yields this value of k_F would then serve to confirm the adequacy of the free electron model. If the effect cannot be observed even under good experimental conditions then it is likely that the Fermi Surface is blurred in the liquid and the free electron model is not a good approximation. Another aspect that makes such an experiment of interest is the possibility of observing the Fermi Surface in liquid metal solutions, especially of metals having different valences. Here again one would like to know whether the Fermi Surface is sharp and whether the free electron model predicts the correct value of k_F .

In this report we present the results of a measurement of the quasi-elastic widths in liquid lead at 350°C in a preliminary search to observe the Kohn effect. According to the free electron model it should occur at $\kappa = 2k_F = 3.08 \text{ \AA}^{-1}$. The results comprise data from two separate experiments. The newer experiment was specifically performed to search for the Kohn effect in the region of $2k_F$, while the data of the earlier one, which has already been reported in another context⁽⁶⁾, extends the measurements over a wider κ range. The older experiment will be called experiment "A" and the newer one will be called experiment "B". After mentioning the common features of experiments A and B, we summarize for convenience some of the pertinent details of A, then go into a more detailed discussion of B.

II. THE EXPERIMENT

Both experiments A and B were performed with the MTR phased chopper velocity selector. The speed of both the choppers and the rotating collimators was 7500 rpm. The sample was in the form of a rectangular slab 11.43 cm high by 7.62 cm wide. The sample holder is in the form of two rectangular steel frames which sandwich the spacer determining the sample thickness and the windows which pass the neutrons. The frames were coated with a paint impregnated with gadolinium oxide powder to reduce unwanted scattering from the frame. An empty sample holder of identical construction was used to obtain background scattering. The sample temperature was $350 \pm 5^{\circ}\text{C}$.

A. Experiment A

This experiment was performed using a helium filled flight path at 15 scattering angles located every 5° between 15° and 90° . The

detectors at each angle were composed of three 18-inch active length BF_3 filled proportional counters of 1-inch diameter. These subtend 1.96° at the sample. The scattered neutron flight path was 2.25 m and a time-of-flight channel width of 10 μsec was used. The sample windows were of 0.0025 cm vanadium and bulging of the windows was reduced by using three horizontal steel support ribs painted with gadolinium oxide powder. The nominal sample thickness was 0.127 cm and the measured sample transmission at 0.02 eV was 0.91. These measurements were made at incident energies of 14.9, 17.0 and 20.0 meV and spanned the κ range $1 \leq \kappa \leq 4.5 \text{ \AA}^{-1}$. The energy resolution for elastic scattering as measured by the beam monitor was $\Delta E \leq 0.8$ meV. Typical wavelength spectra are given in Ref. (4). The quasi-elastic width measurements of the reduced partial differential scattering cross section $S(\kappa, \Delta E)$ obtained at fixed scattering angles span the desired range. However right at $\kappa = 3.08 \text{ \AA}^{-1}$ there were contributions arising from Bragg scattering in the steel support ribs of the sample (iron (111) planes). These did not completely subtract out in the background correction, probably due to a thin spot in the Gd_2O_3 paint on one of the support ribs. This effect gave rise to anomalously narrow quasi-elastic widths at $3.08 \text{ \AA}^{-1}$ and could not be used.

B. Experiment B

This experiment was designed to avoid the Bragg peak problem at $3.08 \text{ \AA}^{-1}$ which occurred in experiment A and at the same time use narrower detectors and smaller channel widths (5 μsec). To eliminate the problem of spurious contribution to the widths arising from Bragg scattering, the same sample holder was used but the cover windows

were of 0.051 cm molybdenum, and no support ribs were needed. The sample was 0.127 cm thick and had a measured transmission of 0.93 at 24.0 meV. The advantage of the molybdenum is that there are no Bragg peaks in the region near $\kappa = 3.08 \text{ \AA}^{-1}$. The nearest ones are at 2.82 and $3.95 \text{ \AA}^{-1}$. Data that did contain these peaks were not used. The sample and the entire scattered neutron flight path were contained in an evacuated flight path. This flight path subtends approximately 20° and was centered at the 54° port of the scattering chamber. Seven scattering angles were located at angles between 45° and 62° . The angles were selected such that at the incident energies used the three detectors in the middle spanned a κ range in the neighborhood of $3.08 \text{ \AA}^{-1}$, while the remaining four were located with two detectors at each end of the angular range. The detectors were each composed of two 6-inch active length, 1-inch diameter He^3 filled proportional counters and subtend 1.28° at the sample. Longer active length would have been preferable, but could not be accommodated by the flight path. The length of the scattered flight path was 2.26 m. Four runs were made at incident energies of 23.25, 24.00, 24.49, and 24.98 meV.

C. Energy and Momentum Resolution

There are two kinds of energy "resolution" effects that influence quasi-elastic width measurements. The first is a true resolution effect arising from the finite spread of energies in the incident beam. This effect broadens the observed spectra. The other is an uncertainty in the value of the measured quasi-elastic width due to the counting statistics on the data points. The first effect or resolution can be obtained from measurements of the width of the time-of-flight burst of the transmitted neutrons as measured at the forward beam monitor.

It gives the resolution for elastic scattering. The fractional time resolution is $\frac{\Delta t}{t}$ where t is the sample-to-monitor time of flight, and $\Delta t = 2\sigma_t$ where σ_t^2 is the variance of the burst at the detector and $\frac{\Delta E}{E} = \frac{2\Delta t}{t}$. Table I summarizes the energy resolution for experiments A and B.

TABLE I
ENERGY RESOLUTION

Experiment	Run Number	E_o (meV)	$2\sigma_t$ (μs)	$\frac{\Delta t}{t}$	$\frac{\Delta E}{E_o}$	ΔE (meV) = $2\sigma_R$
A	1432	14.90	22.66	.0170	.0340	0.506
A	1438	17.00	22.83	.0175	.0350	0.618
A	1446	20.00	22.00	.0191	.0382	0.764
B	1806	24.00	22.16	.0197	.0394	0.946
B	1814	24.50	21.43	.0193	.0386	0.946
B	1820	23.25	23.22	.0203	.0406	0.944
B	1832	25.00	21.65	.0196	.0392	0.980

At small energy transfer the resolution in momentum is dominated by the finite angular spread of the detector. For experiment A the spread is $\Delta\kappa \cong \pm .05 \text{ \AA}^{-1}$, while for experiment B it is $\Delta\kappa \cong \pm .035 \text{ \AA}^{-1}$. There is also a slight uncertainty arising from the fact that the width measurements are made on scattered neutron spectra at fixed scattering angles rather than at constant κ . This generally amounts to $\Delta\kappa \approx .03 \text{ \AA}^{-1}$. The overall κ resolution is therefore taken to be $.05 \leq \Delta\kappa \leq .07 \text{ \AA}^{-1}$.

Figure 1 - Typical wavelength spectrum of raw data for experiment B. The elastic peak in the background is due to a small amount of incoherent scattering in the molybdenum cover plates of the sample.

D. Results and Analysis

Figure 1 shows a typical spectrum of sample and background raw data obtained in experiment B at an angle of 53.9° and an incident energy of 23.25 meV ($\lambda_0 = 1.83 \text{ \AA}$). The elastic peak in the background is incoherent and arises from a small incoherent scattering cross section of the molybdenum cover plates of the sample holders. It is seen from this plot that there is no really clear cut separation of the quasi-elastic scattering from the inelastic scattering. Figure 2 shows the data for one run after correction for background and detector efficiency and is proportional to the cross section $\frac{d^2\sigma}{d\Omega dE}$. The error flags are due to counting statistics only, and the curves are hand drawn through the data. In the region of the full width at half maximum the spectra contain not only the

Figure 2 - Representative cross section data for liquid lead at 350°C. The solid curve is hand drawn and the dotted curves show the limits used to obtain the energy widths. The values of ΔE are the full widths at three-fourths maximum.

"quasi-elastic" contributions of interest, but also effects of energy resolution, inelastic scattering and multiple scattering. This is also true of experiment A. All of these effects tend to broaden the peak. Thus slight changes in the quasi-elastic contribution to this width, such as expected from the Kohn effect tend to be washed out. At smaller energy transfers however the inelastic scattering and multiple scattering contribute less to the spectrum. In order to reduce these extraneous contributions and see the quasi-elastic part more clearly, we have arbitrarily used the widths at 3/4 maximum rather than at 1/2 maximum. The error in the full width at 3/4 of the maximum (FW 3/4M) is estimated by measuring the widths of the peaks obtained by drawing the curves following both the upper and lower ends of the error

flags (Figure 2).

In order to combine the results at the various incident energies we have made an approximate correction for the energy resolution, which changes slightly with the different incident energies (see Table I). The correction is made by assuming that in the neighborhood of the FW 3/4M all line shapes are approximately Gaussian. Then the observed full width at 3/4 maximum W_o , the natural line width at 3/4 maximum W_n , and the resolution function at 3/4 maximum W_R are related by

$$W_n^2 = W_o^2 - W_R^2$$

For a Gaussian resolution function the relation between the standard deviation $\Delta E = 2\sigma_R$ of the resolution function as given in Table I and W_R the FW 3/4M is

$$W_R = 0.759 \Delta E$$

This correction was made on all measured widths. After making this correction a normalization of 1.15 was needed to bring the results of experiment A into agreement with that of B in the κ region where they overlap. This probably means that the assumption of Gaussian line shapes is not very good. However since the point of interest is the variation of the FW 3/4M with κ rather than the absolute value of the width, the use of the normalization factor is reasonable.

Figure 3 shows the energy widths FW 3/4M obtained from the two experiments and for comparison also shows the diffraction pattern for liquid lead. The widths have been corrected for resolution and the A experimental widths have been normalized by the above mentioned factor. The κ values assigned are those at the maximum in the spectrum of each scattering angle. The points have a scatter of about 10%

Figure 3 - Plot of the full widths at three-fourths maximum obtained at fixed scattering angle from experiments A and B.

over most of the κ range but is larger at both the highest and lowest values. No abrupt slope discontinuity can be seen at $\kappa = 3.08 \text{ \AA}^{-1}$. Therefore if the Kohn effect does exist in liquid lead as a sharp slope change at $\kappa = 2k_F$, we have been unable to observe it due to the scatter of the data. The main features of the width curve is that the widths show oscillations around a general increase with increasing κ . The lines are narrowest at $\kappa \approx 2.0 \text{ \AA}^{-1}$, show a local maximum at $\kappa \approx 3.2 \text{ \AA}^{-1}$ and a flattening or perhaps a local minimum at $\kappa \approx 3.7 \text{ \AA}^{-1}$. This oscillation of the line width was predicted by DeGennes⁽⁷⁾ and is a general property of the coherent scattering in liquids. The general feature of DeGennes' calculations is that the quasi-elastic peak will show a narrowing wherever the diffraction pattern has a maximum and line broadening where the pattern has a minimum. Examining the diffraction pattern in Figure 3 the line narrowing in the region of the first diffraction maximum is observed, and the line then broadens with increasing κ and has a maximum at $\kappa \approx 3.2 \text{ \AA}^{-1}$ where the diffraction pattern has a minimum. So far the variations of the line widths agree with DeGennes. However in the region of the second diffraction maximum, where a line narrowing would again be expected ($\kappa \approx 4.2 \text{ \AA}^{-1}$) the line is simply increasing in width. Also at $\kappa \sim 3.0 \text{ \AA}^{-1}$ there appears to be a flattening or perhaps a slight minimum in the line width curve. These observations are borne out from width measurements made earlier in experiment A which are shown in Figure 4. In these measurements the full width at half maximum was obtained and no correction for resolution ($W_R = 0.80 \text{ meV}$) were made. The widths were obtained from the scattering law $S_R(\kappa, \omega)$ (Figure 6 of Ref. 6) at selected constant values of κ , rather than at constant scattering

Figure 4 - Plot of the full widths at one-half maximum obtained at constant κ from plots of the scattering law $S(\kappa, \omega)$ in experiment A(6).

theoretical curve is that given by Vineyard⁽¹⁰⁾ for particles which undergo simple diffusive motion. Figure 4 extends over a wider range of κ and shows that there is no line narrowing at the second diffraction maximum. The line widths are in good agreement with those of Egelstaff below $2.5 \text{ \AA}^{-1}$ but above that are consistently narrower. It is clear that this discrepancy cannot be due to energy resolution broadening since Egelstaff corrected for resolution in Brockhouse's data, whereas our data of Figure 4 has not been corrected for resolution.

angle, so that the values of ΔE from Figures 3 and 4 should not be directly compared. We note however that the general shape is quite similar, in fact the FW 1/2M curves of Figure 4 show a more pronounced minimum at $\kappa \approx 3.6 \text{ \AA}^{-1}$ than the FW 3/4M curves of Figure 3. We see that at $\kappa = 2k_F$ there is no apparent abrupt slope change, in agreement with the newer results. Also shown in Figure 4

are the line width results of Egelstaff⁽⁸⁾ and those of Brockhouse and Pope⁽⁹⁾ as analyzed by Egelstaff⁽⁸⁾. The solid

III. CONCLUSIONS

1) The Kohn anomaly has not been observed in liquid lead; the data is of insufficient precision to see it if the anomaly occurs as an abrupt slope change at the free electron value of $2k_F$. The line widths curve changes its slope continuously in this region. It is possible however that liquid lead is not the best material in which to attempt this measurement. Enderby and March⁽¹¹⁾ have made estimates of the spread in k , or the electron coherence range, for liquid lead near k_F . This spread Δk gives a measure of the fuzziness of the Fermi Surface. For liquid lead they estimate a value of $\frac{\Delta k}{k_F} = 0.11$. Thus the surface may not be sharp, so that the Kohn anomaly would either not occur at all or else occur as a gradual slope change indistinguishable from the changing of the slope due to other causes. However similar estimates for liquid sodium give a value of $\frac{\Delta k}{k_F} = 0.007$, so that sodium may be a much better material in which to look for the anomaly.

2) The κ position for which the quasi-elastic line width narrows or broadens is in agreement with DeGennes at the first maximum and first minimum of the diffraction pattern, but does not agree with his predictions at higher κ values.

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