[17]I. P. Suzdalev, M. Ya. Gen, V. I. Goldanskii, and E. F. Makarov, Zh. Eksperim. i Teor. Fiz. 51, 118 (1966) [translation: Soviet Phys.-JETP 24, 79 (1967)].
[18]C. P. Bean and J. D. Livingston, J. Appl. Phys. Suppl. 30, 120S (1959).
[19]K. J. Ando, W. Kündig, G. Constabaris, and R. H. Lindquist, J. Phys. Chem. Solids 28, 2291 (1967).
[20]A. M. Van der Kraan and J. J. Van Loef, Phys. Letters 20, 614 (1966).
[21]W. J. Schuele, S. Shtrickman, and D. Treves, J. Appl. Phys. 36, 1010 (1965).
[22]C. J. Coston, R. Ingalls, and H. G. Drickamer, Phys. Rev. 145, 409 (1966).
[23]Handbook of Physical Constants (Geological Society of America, New York, 1942).
[24]J. S. Smart, in Magnetism, edited by G. T. Rado and H. Suhl (Academic Press, Inc., New York, 1963), Vol. III, p. 63f.
[25]C. Kittel, Introduction to Solid State Physics (John Wiley & Sons, Inc., New York, 1967), 3rd ed., p. 455f.
[26]V. G. Bhide and G. K. Shenoy, Phys. Rev. 143, 309 (1966).
[27]J. T. Richardson and W. O. Milligan, Phys. Rev. 102, 1289 (1956).

OBSERVATION OF LOCALIZED VIBRATIONS IN Cu-4% Al BY COHERENT INELASTIC NEUTRON SCATTERING*

R. M. Nicklow, P. R. Vijayaraghavan, H. G. Smith, and M. K. Wilkinson
Solid State Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee
(Received 4 March 1968)

The localized vibrations of Al in a Cu host lattice have been observed in a Cu-4% Al crystal by coherent inelastic neutron scattering. Both the local-mode frequency and the $\vec{q}$ dependence of the neutron scattering cross sections are in good agreement with a calculation based on a mass-defect theory.

In this Letter we report some preliminary results of coherent inelastic neutron scattering measurements of the localized vibrational modes in a Cu single crystal containing 4.1 at.% Al as a substitutional impurity. The experiments were performed on a triple-axis neutron spectrometer at the High Flux Isotope Reactor (HFIR). All the measurements were obtained with the spectrometer operating in the constant-$\vec{Q}$ mode[1] and with the scattered neutron energy E' held fixed at 18.9 meV (4.5×10^{12} cps). Inelastic-neutron-scattering studies of localized vibrations have been reported previously,[2-4] but to our knowledge there have been no previous coherent-inelastic-scattering measurements of local vibrational mode in single crystals.

The cross section for the coherent inelastic neutron scattering from a concentration c of isolated light-mass substitutional impurities in a cubic lattice, provided there is no change in the interatomic force constants, is given by Elliott and Maradudin[5] as

$$\frac{d^2\sigma}{dEd\Omega}=\frac{k_0}{2k'}(n_l+1)\delta(\nu-\nu_l)\frac{cb^2\nu_l}{MB(\nu_l^2)}e^{-2W}\sum_j[\vec{Q}\cdot\vec{e}_j(q)]^2\left[\frac{b'/b-1}{\nu_l^2\epsilon}+\frac{1}{\nu_l^2-\nu_j^2(q)}\right]. \quad (1)$$

Only energy loss of the neutron is considered. n_l is the equilibrium number of phonons with frequency ν_l, e^{-2W} is the Debye-Waller factor, and b' and b are the scattering lengths of the impurity and host, respectively. $\vec{Q}$ is the wave-vector transfer of the neutron, $\vec{e}_j(\vec{q})$ and $\nu_j(\vec{q})$ are the polarization vector and frequency of the normal mode in the pure crystal with wave vector $\vec{q}$ and branch index j, and $\epsilon=1-(M'/M)$, where M' is the impurity mass and M is the host mass. ν_l is given by the solution of

$$\nu_l^2\int\frac{g(\nu)d\nu}{\nu_l^2-\nu^2}=\frac{1}{\epsilon}, \quad (2)$$

where $g(\nu)$ is the frequency distribution function (normalized to unity) of the unperturbed host lattice, and

$$B(\nu_l^2)=\int\frac{\nu^2g(\nu)d\nu}{(\nu_l^2-\nu^2)^2}. \quad (3)$$

The concentration of impurities in the sample studied in the present work may not be sufficiently small for each impurity to be considered isolated. In addition, one might expect changes in the interatomic force constants, since there is a tendency toward short-range order in the more concentrated Cu-Al alloys.[6] Nevertheless, Eqs. (1)-(3) provide a surprisingly good prediction of the observations made in the present study.

In Fig. 1 are shown identical constant-$\vec{Q}$ scans for pure Cu and Cu-4% Al at two points in reciprocal space. For pure Cu only the expected peaks due to the $L(\zeta\zeta\zeta)$ phonons appear. The zone-boundary phonon [Fig. 1(a)] in this symmetry direction is the highest frequency phonon [$(7.36\pm0.08)\times10^{12}$ cps] that is observed in Cu. In Cu-4% Al the $L(\zeta\zeta\zeta)$ phonon peaks are shifted to lower frequencies and an additional peak is observed at $(8.78\pm0.1)\times10^{12}$ cps. This additional peak is interpreted as neutron scattering by the high-frequency, localized vibrations of Al in the Cu host lattice. The value for the local-mode frequency ν_l calculated with Eq. (2) and a $g(\nu)$ for Cu recently reported[7] is 8.48×10^{12} cps, which is in quite good agreement with experiment. The discrepancy between the calculated and measured ν_l, though small, is felt to be significant and is presumably due to small force-constant changes and/or effects of the high Al concentration. The observed energy width of the local-mode scattering is not significantly different from the zone-boundary phonon of Cu or of Cu-Al and, at present, can be accounted for entirely by the instrumental resolution.

From a comparison of Eq. (1) [for $B(\nu_l^2)\simeq0.02$] with the one-phonon coherent cross section,[8] one finds that the local-mode scattering intensity at $\zeta=0.50$ is predicted to be about 15% of that for the $L(0.5,0.5,0.5)$ phonon. This result is consistent with the observed relative intensities. Figures 1(a) and 1(b) reveal a reduction in the local-mode peak intensity as $\vec{Q}$ moves from the zone boundary. Several constant-$\vec{Q}$ scans performed to investigate this intensity variation are shown in Fig. 2. These scans clearly show that the intensity decreases as $\vec{Q}$ moves away from the $(\zeta\zeta\zeta)$ zone boundary, and such a decrease is predicted by Eq. (1). Because $\vec{Q}$ lies along the [111] direction in these experiments, only the longitudinal branch contributes to the sum over j, and at the zone boundary the $L(\zeta\zeta\zeta)$ branch reaches its highest frequency. Therefore, the second term in the square brackets in Eq. (1), and hence the cross section, is a maximum there. The first term in the square brackets is independent of $\vec{Q}$ and approximately equal to $-1/\nu_l^2$ for Al in Cu. A comparison of the $\vec{Q}$ dependence of the relative intensity of the peaks in Fig. 2 with that predicted in Eq. (1) is shown in Fig. 3. The errors shown on the experimental points are based on

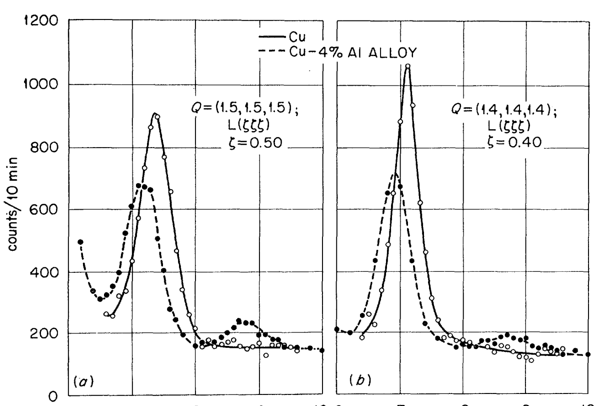

FIG. 1. Scattered intensity versus frequency for constant-Q scans at the $(\zeta\zeta\zeta)$ zone boundary in Cu and Cu-4% Al. The $L(\zeta\zeta\zeta)$ zone-boundary phonons as well as the local mode in Cu-4% Al are observed in these scans.

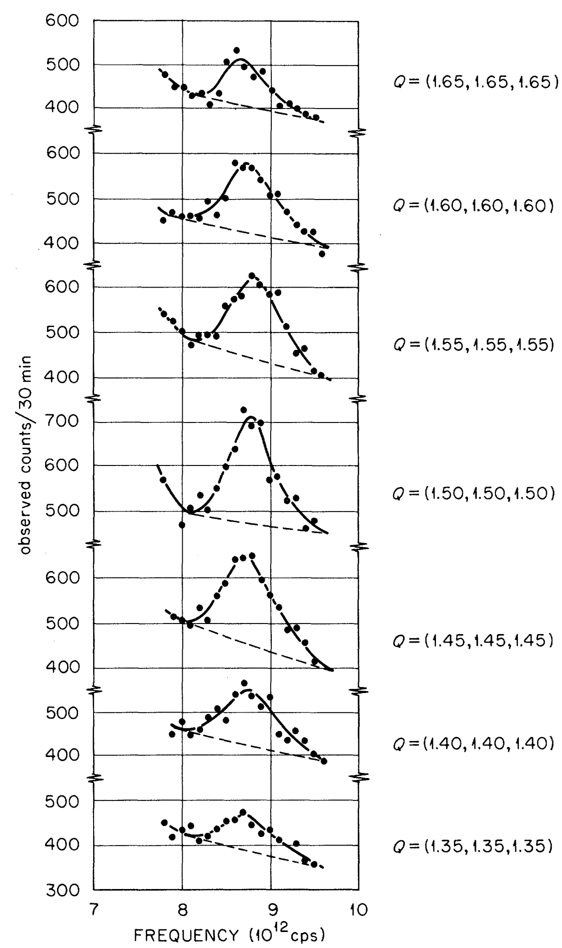

FIG. 2. Several constant-Q scans at and near the $(\zeta\zeta\zeta)$ zone boundary in Cu-4% Al, illustrating the Q dependence of the local-mode scattering intensity.

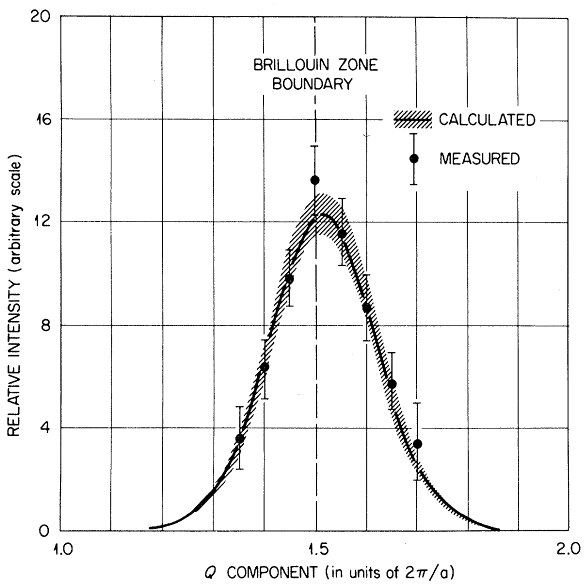

FIG. 3. Comparison of the observed Q dependence of the local-mode scattering intensity with theory, Eq. (1).

counting statistics only. The shaded region around the calculated curve is an estimate of the uncertainty in the calculations arising from the experimental errors in $\nu_j(\vec{q})$ and ν_l. The amplitude of the calculated curve has been adjusted so that the curve passes through the experimental point at $\vec{Q}=(1.55,1.55,1.55)$. The fit to the rest of the data is exceedingly good.

It should be pointed out that although the data shown in Fig. 2 have not been corrected to an absolute intensity scale, such corrections would have been independent of $\vec{Q}$. Since the local-mode frequency is independent of $\vec{Q}$, the incident neutron energy E_0 was varied over the same range for all the scans. However, the $1/k_0$ dependence of the neutron scattering cross section, Eq. (1), was largely compensated experimentally by the k_0 (or $1/v_0$, v_0 being the neutron velocity) dependence of the efficiency of the monitor counter placed in the beam before the sample (see Ref. 8). Also, because the sample was a cylinder which had its axis oriented nearly perpendicular to both the incident and scattered neutron beams, the sample volume intercepted by the beam did not vary from scan to scan. Therefore, a comparison of the relative intensities of these local-mode scattering peaks could be made without the application of corrections to the data other than the background subtractions indicated by the dotted lines in Fig. 2.

The $\vec{Q}$ dependence of the neutron cross section can be related to the spatial distribution of the vibrational displacements in the vicinity of the impurity atom. Since the theory is in excellent agreement with the measured $\vec{Q}$ dependence of the cross section, we have related the Fourier transform of the second term in the square brackets in Eq. (1) to the asymptotic decay of the localized mode, as suggested by Maradudin[9] (see also Ref. 5). Following Maradudin[9] we make the following approximation:

$$\nu_l^2-\nu^2(\vec{q})\approx\nu_l^2-\nu_m^2+\tfrac{1}{2}A_{111}|\vec{q}-\vec{q}_m|_{111}^2, \qquad (4)$$

where ν_m and $\vec{q}_m$ are the frequency and wave vector for the maximum of the $L(\zeta\zeta\zeta)$ branch. For the range of $\vec{q}$ studied in the present experiment, this approximation with $A_{111}\simeq 6.9a_0$ (a_0 is the lattice parameter for Cu) agrees within ±1% with the dependence of $\nu_l^2-\nu_j^2(\vec{q})$, obtained from the experimental values for ν_l and $\nu_j(\vec{q})$. The above approximation then leads to an asymptotic decay of the local-mode displacement, which varies exponentially with distance from the impurity atom, with an inverse decay length in the (111) direction of

$$[2(\nu_l^2-\nu_m^2)/A_{111}]^{1/2}\simeq 2.6/a_0. \qquad (5)$$

The analysis indicates that the local mode in Cu-4% Al is extremely well localized.

This study is being extended to other crystallographic directions in Cu-4% Al and to several other Al concentrations, and investigations of the "in-band" modes are also underway. Preliminary results on both a Cu-4% Al and a Cu-10% Al crystal show changes in the frequencies of the "in-band" modes, which are in poor agreement with theory, in contrast to the good agreement found in this study of local modes. For example, although the mass-defect theory[5] does predict a decrease in the frequency of the zone-boundary $L(\zeta\zeta\zeta)$ mode, in qualitative agreement with the observation shown in Fig. 1(a), the observed decrease is almost twice that calculated. Since the frequencies measured for the in-band modes and the local modes depart from the theoretical predictions in opposite directions, presumably a consideration of both force-constant changes and concentration effects will be required to bring the theory into better agreement with the experimental results. A more detailed discussion of the comparison of theory with the experimental measurements of the in-band modes will be presented in a future publication.

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[1]B. N. Brockhouse, Inelastic Scattering of Neutrons in Solids and Liquids (International Atomic Energy Agency, Vienna, 1961), p.113.

[2]I. Natkaniec, K. Parlinski, A. Bajorek, and M. Sudnik-Hrynkiewicz, Phys. Letters 24A, 517 (1967).

[3]B. Mozer, V. W. Muers, and K. Otnes, Phys. Rev. Letters 8, 278 (1962); see also B. Mozer, K. Otnes, and C. Thaper, Phys. Rev. 152, 535 (1966).

[4]M. G. Zemlyanov, V. A. Somenkov, and N. A. Cheroplenkov, Zh. Eksperim. i Teor. Fiz. 52, 665 (1967) [translation: Soviet Phys.–JETP 25, 436 (1967)].

[5]R. J. Elliott and A. A. Maradudin, Inelastic Scattering of Neutrons in Solids and Liquids (International Atomic Energy Agency, Vienna, 1965), Vol. I, p. 231.

[6]Bernard Borie and C. J. Sparks, Jr., Acta Cryst. 17, 827 (1964).

[7]R. M. Nicklow, G. Gilat, H. G. Smith, L. J. Raubenheimer, and M. K. Wilkinson, Phys. Rev. 164, 922 (1967).

[8]B. N. Brockhouse, L. N. Becka, K. R. Rao, and A. D. B. Woods, Inelastic Scattering of Neutrons in Solids and Liquids (International Atomic Energy Agency, Vienna, 1963), Vol. II, p. 23.

[9]A. A. Maradudin, Solid State Phys. 18, 273 (1966).

PROPERTIES OF THE NEUTRONS EMITTED IN THE LONG-RANGE ALPHA FISSION OF Cf^{252}

Eran Nardi
Israel Atomic Energy Commission, Soreq Research Center, Yavne, Israel
and Weizmann Institute of Science, Rehovoth, Israel

and

Zeev Fraenkel
Weizmann Institute of Science, Rehovoth, Israel
(Received 25 March 1968)

We have investigated the long-range alpha (LRA) fission neutrons of Cf^{252} in a four-parameter correlation experiment and found the behavior of the average number of neutrons as a function of fragment mass and total fragment kinetic energy to be very similar in binary and LRA fission. We concluded that binary and LRA fission have essentially the same scission configuration and discuss the mechanism of alpha emission and the fragment stiffness coefficients also.

Neutrons emitted in the spontaneous fission of Cf^{252} accompanied by long-range α particles (LRA) particles have been investigated in a four-parameter correlation experiment. A schematic representation of the experiment is presented in Fig. 1. Four fixed solid-state counters, two for fission-fragment detection and two for α-particle detection, were placed inside an aluminum vacuum chamber of 30-cm diam and 5-mm wall thickness. The fission counters were locally made 300-Ω-cm surface-barrier detectors. Both detectors subtended an angle of 44° with respect to the source. The α-particle detectors were surface-barrier counters, manufactured by ORTEC, of 300-μ depletion layer. They were placed opposite each other at 90° to the fission-counter axis and each subtended an angle of 60° with respect to the source. The source consisted of Cf^{252} on a 100-$\mu g/cm^2$ Ni backing. Its strength was about 2×10^5 fissions/min. The source plane was at 45° to the four detectors. In order to prevent fission fragments and 6.1-MeV α particles from the Cf^{252} α decay from reaching the α-particle detectors, 17-mg/cm^2 gold foils were placed between the Cf^{252} source and the α counters.

The neutrons were detected by means of the time-of-flight method with the aid of two identical NE102 plastic scintillators of 5 in. diam and

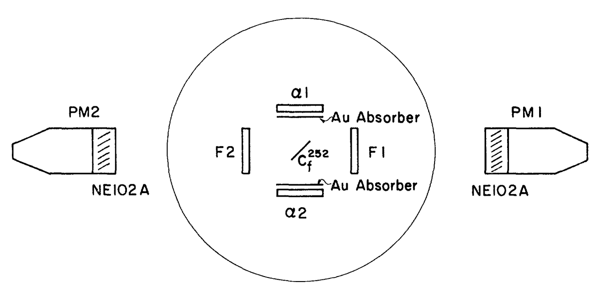

FIG. 1. Experimental arrangement.