garded as additional to normal band effects in metals, discussed e.g., by Haque and Kliewer.[12]

The rather small gap values we find for simple metals can explain the apparent absence of a gap in the experiments performed so far. We find however that the effect should be of observable magnitude, at least with modulation techniques, in Sn and Ge. More generally, Landau damping and nonlocality tend to quench the plasmon gap at $G/2$ as soon as $G/2 > q_c$ and $v_G^{\text{atomic}} > 0$, so that good candidates are substances with $G/2 < q_c$ (narrow plasmons) and $v_G^{\text{atomic}} < 0$. Further extensions of this calculation could include band effects of order higher than v_G,[10] non-RPA exchange and correlation processes,[13] and a study of the plasmon bands over the entire Brillouin zone.

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[1]N. H. March and M. P. Tosi, Proc. Roy. Soc., London A330, 373 (1972).

[2]W. M. Saslow and G. F. Reiter, Phys. Rev. B 7, 2995 (1973).

[3]K. C. Pandey, P. M. Platzman, P. Eisenberger, and E.-Ni Foo, Phys. Rev. B 9, 5046 (1974).

[4]D. M. Miliotis, Phys. Rev. B 3, 701 (1971); P. Eisenberger, P. M. Platzman, and K. C. Pandey, Phys. Rev. Lett. 31, 311 (1973); P. M. Platzman and P. Eisenber-

ger, Phys. Rev. Lett. 32, 152 (1974); P. Eisenberger, P. M. Platzman, and P. Schmidt, Phys. Rev. Lett. 33, 18 (1975).

[5]P. Zacharias, J. Phys. C 7, L26 (1974); H. J. Höhberger, A. Otto, and E. Petri, Solid State Commun. 16, 175 (1975); P. Zacharias, J. Phys. F 5, 645 (1975); P. C. Gibbons, S. E. Schnatterly, J. J. Ritsko, and J. R. Fields, to be published.

[6]C. M. Bertoni, V. Bortolani, C. Calandra, and F. Nizzoli, J. Phys. F 4, 19 (1974).

[7]C. J. Powell, Proc. Phys. Soc., London 76, 593 (1960).

[8]D. Pines, Elementary Excitations in Solids (Benjamin, New York, 1963), Chap. 4.

[9]The opening and closing of such umklapp damping channels may well be responsible for some of the observed q dependence of ω_I, as measured in several metals [see P. Zacharias, Z. Phys. 256, 92 (1972); T. Kloos, Z. Phys. 265, 225 (1973)].

[10]Calculations of plasmon damping and changes of dispersion caused by the periodic lattice structure have been performed by several authors. See G. Paasch, Phys. Status Solidi 38, K123 (1970); M. Hasegawa, J. Phys. Soc. Jpn. 31, 649 (1971); M. S. Haque and K. L. Kliewer, Phys. Rev. B 7, 2416 (1973). Plasmon gaps at $q = G/2$ are not involved in these works, either because they restrict to small q (Paasch and Hasegawa), or because Umklapp response (i.e., local-field corrections) is neglected (Haque and Kliewer).

[11]Zacharias, Ref. 9; Kloos, Ref. 9.

[12]Haque and Kliewer, Ref. 10.

[13]D. F. Dubois and M. G. Kivelson, Phys. Rev. 126, 409 (1969).

Measurement of the Mobility of Positrons in Germanium

Allen P. Mills, Jr., and Loren Pfeiffer

Bell Laboratories, Murray Hill, New Jersey 07974

The mobility μ_+ of positrons in Ge has been measured. Energetic positrons from a radioactive source enter an intrinsic planar Ge detector where they annihilate in the presence of the detector bias field. The motion of the positrons is detected by observing the Doppler shift of the annihilation γ's as a function of the voltage across the detector. We obtain $\mu_+ = (124 \pm 10)$ cm^2 V^{-1} sec^{-1} at (93 ± 5) K and $\mu_+ = (350 \pm 17)$ cm^2 V^{-1} sec^{-1} at (36 ± 5) K.

In this Letter we report the first measurement of a positron mobility μ_+ in a solid. The experiment was motivated by a desire to understand the mechanism whereby the energetic positrons from a radioactive source are converted to a low-energy (few eV) beam by means of a solid moderator.[1] One of the parameters needed for such understanding is the diffusion constant for positrons D_+ which may be obtained from the mobility

using the Nernst–Einstein relation $D_+ = \mu_+ kT/e$. Knowledge of D_+ also might be useful in the absolute determination of vacancy concentrations by positron-annihilation techniques.[2] The positron mobility is of further interest because from it the positron thermalization time[3] due to acoustic-phonon scattering in insulators or semiconductors can be estimated.

The mobility of a positron in a solid is defined

by the relation $\vec{v}_+ = \mu_+ \vec{\mathscr{E}}$, where $\vec{\mathscr{E}}$ is the electric field in the solid and $\vec{v}_+$ is the positron drift velocity. If the electrons in the solid are immobile, the positron drift velocity at the time of annihilation may be obtained from a measurement of the center-of-mass momentum $m\vec{v}_+$ by observing the annihilation photons and using for m the free positron mass. The momentum distribution of the annihilating pair from which we may find $m\vec{v}_+$ may be obtained either from a study of the 2γ angular correlation or from a measurement of the 2γ Doppler-shift distribution.[4] Thus if the electric field $\vec{\mathscr{E}}$ is parallel to the momentum of the annihilation photon, there will be a Doppler shift of the mean photon energy E given by $\Delta E/E = v_+/2c$. In an angular-correlation experiment, the centroid of the angular distribution would shift an amount $\Delta\theta = mv_+c/E$ if $\vec{\mathscr{E}}$ is perpendicular to the direction of emission of the two photons. The expected shifts caused by the positron drift velocity ($\sim 10^6$ cm sec^{-1}) are small compared to the widths of these distributions which arise primarily from the electron momenta corresponding to velocities $\sim \alpha c \approx 2\times 10^8$ cm sec^{-1}.

The first attempt to detect the mobility of positrons in a solid was an angular-correlation measurement on diamond at room temperature by Lang and DeBenedetti[5] which yielded the result $\mu_+ = c\Delta\theta/\epsilon = (120\pm 160)$ cm^2 V^{-1} sec^{-1}. More recently this measurement was repeated by Sueoka and Koide[6] who found $\mu_+ < 20$ cm^2 V^{-1} sec^{-1}. The same authors also found $\mu_+ < 15$ cm^2 V^{-1} sec^{-1} for SiO_2 and $\mu_+ < 80$ cm^2 V^{-1} sec^{-1} in $BaTiO_3$. There is in addition to the above one Doppler-shift experiment using a coaxial Ge(Li) detector by MacKenzie and Campbell.[7] These workers found a 50-eV shift in the single escape peak when the detector bias was changed by 600 V. The origin of this shift is unclear because the mobility one estimates from this measurement would be ~ 40 times larger than expected from the results reported here.

It can be inferred from diffusion experiments using powdered samples that positrons could have an appreciable mobility in some materials. For example, measurements of the positron-annihilation lifetime in powdered metals at 300 K allowed Paulin, Ripon, and Brandt[8] to estimate $D_+ = (1\pm 0.5)\times 10^{-2}$ cm^2 sec^{-1}. The Einstein relation then implies $\mu_+ = (0.4\pm 0.2)$ cm^2 V^{-1} sec^{-1} for these samples. Similar lifetime measurements[9] on powdered Ge and Si indicate that D_+ could be much larger in a semiconductor.

In a metal a positron is expected to have a low

mobility[10] owing to positron collisions with electrons near the Fermi energy. In a semiconductor or insulator where the carrier density is low the mobility should be limited (1) by impurity and crystal-defect scattering, (2) by trapping of the positron into nonmobile states, (3) by the finite lifetime $\tau_+ \approx 2\times 10^{-10}$ sec of the positron in the solid, and (4) by phonon generation and positron-phonon scattering. The low-electric-field acoustic-phonon-limited mobility is predicted to be[11]

$$\mu_+ = [(8\pi)^{1/2}/3]v_l{}^2\hbar^4\rho m_+{}^{-5/2}e\epsilon_d{}^{-2}(kT)^{-3/2},$$

where ρ is the density, v_l is the longitudinal sound velocity, and ϵ_d is the energy parameter which characterizes the positron-phonon coupling (the deformation potential). At high electric fields, positrons should have a limiting velocity at about half the velocity threshold for optical-phonon generation as is observed for electrons.[12] It must be remembered that not all positrons in a solid are expected to be mobile. Positronium, known to be formed in many solids, is a zero-mobility state. In addition, positrons can be trapped by impurity centers and a positron could have a very large effective mass and hence a low mobility if it existed in a polaronlike state.[13]

We chose for our sample a Ge single crystal in the form of an intrinsic (IG) planar γ-ray detector.[14] Since such crystals are made from the best high-purity semiconductor material available[15] we hoped that our results would be characteristic of phonon scattering and not masked by the effects of impurities. Other reasons for choosing Ge as a sample are (1) positronium is not observed in Ge,[16] and (2) the effective mass m_+ is $\approx m$.[16]

In our experiment, a ≈ 10-μCi ^{68}Ge source is mounted on a thin Be entrance window 0.4 cm from the front surface of the IG detector (1.1-cm-diam$\times$ 0.5-cm-deep active volume). The 1.8-meV endpoint positrons from the source enter the detector, giving an event-signature pulse from each positron that slows down in the active volume. This pulse is used as a gate to examine the subsequent annihilation γ's in a second and much larger coaxial IG detector (4.3 cm o.d.$\times$ 0.8 cm i.d.$\times$ 2.4 cm deep) located 2.6 cm from the center of the front active surface of the small detector (see insets Fig. 2). The drift velocity of the positron under the influence of the bias field in the small detector appears as a Doppler shift in the 511-keV full-energy peak measured by the large detector. The energy distribution of these γ's is recorded in a 4096-channel analyzer
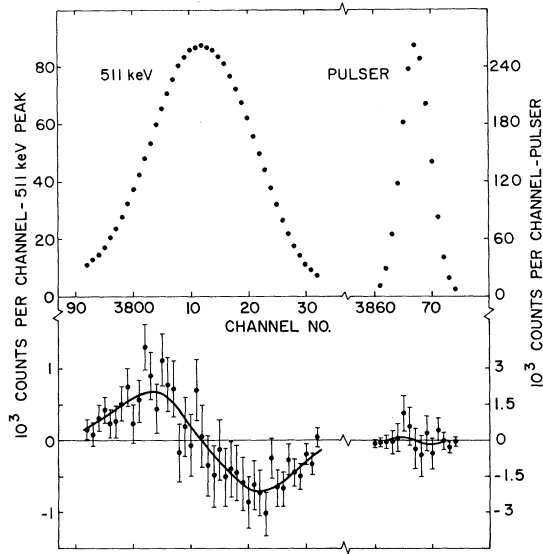
FIG. 1. The 511-keV γ-ray energy spectrum and pulser spectrum for bias $V_2 = 300$ V summed over forty 1-ksec LH_2 runs. The lower portion of the figure shows the channel by channel differences of the $V_2 - V_1 = 3000$ V peaks suitably area normalized. The curves are the first derivatives of the $V_1 + V_2$ summed spectra fitted to the differences using a variable amplitude δ to obtain the 511-keV peak shift (0.1134 ± 0.0103 channels, $\chi^2/\nu = 29.2/40$), the pulser shift (0.0021 ± 0.0025 channels, $\chi^2/\nu = 8.5/14$), and a net Doppler shift of 0.111 ± 0.010 channels out of 3812. The change in velocity inferred for the positrons as $V_2 \rightarrow V_1$ is $(1.76 \pm 0.18) \times 10^6$ cm sec^{-1} toward the large detector.
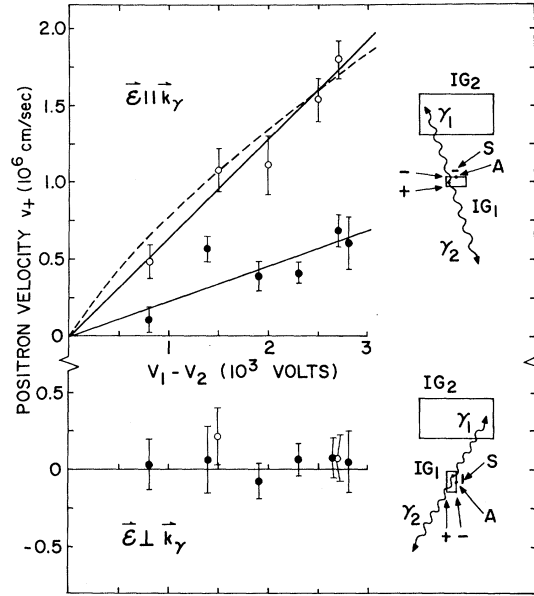

FIG. 2. The change in the positron drift velocity v_+ as a function of bias $V_1 - V_2$ on the 0.5-cm-thick Ge detector sample. This velocity v_+ was obtained from Doppler energy shift measurements using the large detector. Sample temperatures were (36 ± 5) K, open circles, and (93 ± 5) K, solid circles. The insets show the arrangement of the two detectors with $\vec{\mathcal{E}}$ parallel to the momentum of the annihilation γ $\vec{k}_\gamma$ (data and inset in upper portion of figure), and $\vec{\mathcal{E}}$ perpendicular to $\vec{k}_\gamma$ (lower portion). The fitted lines (solid) were constrained to go through the origin. The dashed line is an estimated saturating mobility curve with an initial slope obtained by extrapolating the LN_2 slope to LH_2 by $T^{-3/2}$.

(Northern TN720) as a function of the bias V on the small detector along with the distribution of pulses from a stable reference pulser applied to the preamplifier test input of the large detector. The bias voltage V is applied across the 0.5 cm depth of the small detector ($\vec{\mathcal{E}}$ along the [100] direction). In order to minimize the effects of drift, V is switched between two values, V_1 and V_2, every 1000 sec in an eight step (1, 2, 2, 1, 2, 1, 1, 2) sequence. The spectrum is printed out at the end of each 1000 sec interval.

Figure 1 shows a 511-keV energy spectrum for a given bias V_2 and the accompanying reference pulser spectrum summed over a 1-day run. When the bias is changed to V_1 the 511-keV curve shifts because of the Doppler effect arising from the changed drift velocity of the positrons. The Doppler shift $\delta = \Delta E/E = v_+/2c$ is obtained by fitting each curve by a Gaussian and taking the difference of the fitted centroids. The pulser peak was used to monitor gain drifts of the system which were typically ~ 3 ppm/ksec. Although the sys-

tem is quite stable, residual effects of gain drift are reduced by subtracting the pulser centroid shift from the Doppler shift measured using the 511-keV centroid. Another way of obtaining the shift is to subtract the two area-normalized curves point by point, fitting the result with the first derivative of the sum of the two spectra using δ as a multiplicative parameter. (See the lower portion of Fig. 1.) The two methods are in statistical agreement.

Data were obtained over ~ 100 days for different values of V_1 and V_2. Two sample temperatures were achieved by filling the small-detector Dewar with liquid nitrogen (LN_2) or with liquid hydrogen (LH_2). Figure 2 shows the measured positron drift velocity at the two temperatures plotted against the difference between the two voltages applied to the small detector, $V_1 - V_2$. For most of the measurements V_2 was 300 V [17]; the data are consistent with a linear dependence

on $V_1 - V_2$. The positron mobility is proportional to the slope of the data and is seen to increase as the temperature is lowered, in qualitative agreement with Eq. (1). The sample temperatures [(36 ± 5) K at LH_2 and (93 ± 5) K at LN_2] were found by measuring the temperature at the IG-crystal mount in a dummy detector cryostat of the same model as the one which was used for most of the measurements.

As a control experiment the planar detector was rotated 90° about the point "A" (see insets Fig. 2) so that the bias field was perpendicular to the momentum of the detected 511-keV γ. These $\vec{\mathscr{E}} \perp \vec{k}_\gamma$ points plotted below the $\vec{\mathscr{E}} \parallel \vec{k}_\gamma$ data in Fig. 2 show no Doppler shift as expected. Since nothing is changed in the control experiment except the angle between the two detectors, the null result at 90° is a sensitive check of systematic error.

From the slopes in Fig. 2 (multiplying by a solid-angle correction factor of 1.09) we obtain $\mu_+ = (124 \pm 10)$ cm^2 V^{-1} sec^{-1} at (93 ± 5) K ($\chi^2/\nu = 14.1/5$) and $\mu_+ = (350 \pm 17)$ cm^2 V^{-1} sec^{-1} at (36 ± 5) K ($\chi^2/\nu = 2.1/4$). A similar fit to the $\vec{\mathscr{E}} \perp \vec{k}_\gamma$ data yields a slope consistent with zero as observed above, (9 ± 12) cm^2 V^{-1} sec^{-1} ($\chi^2/\nu = 2.1/7$). The LN_2 point at $V_1 - V_2 = 1400$ V is $\sim 3\sigma$ above the fitted line. The possibility that the point is attributable to nonlinear behavior is being investigated. In addition to the data shown in Fig. 2 an earlier independent set of measurements at LN_2 using three different detector samples and a different less-stable analyzer gave a result consistent with our later measurements, $\mu_+ = (117 \pm 20)$ cm^2 V^{-1} sec^{-1} at (93 ± 5) K ($\chi^2/\nu = 12.3/5$).

Shullman, Beardsley, and Berko[16] have obtained an experimental estimate for m_+ which is consistent with $m_+ \approx m$. Assuming $m_+ = m$ we can use Eq. (1) and our value of μ_+ at 93 K to find $\epsilon_d \approx 19$ eV. This can be compared with the value $\epsilon_d \approx 5$ to 10 eV for holes in Ge.[18] Inasmuch as the deformation potentials calculated in this way for positrons and holes in Ge are in approximate agreement, we can conclude that positrons and holes move in a similar way under the influence of electric fields in Ge crystals after accounting for the difference in their effective masses. The ratio of mobilities observed at the two temperatures $\mu_+(36 \text{ K})/\mu_+(93 \text{ K}) = 2.8 \pm 0.3$ is not as large as predicted by Eq. (1), $(93/36)^{3/2} = 4.2 \pm 0.5$. This may be due to the effects of optical-phonon generation. Indeed the LH_2 data could be consistent with a saturating mobility curve (see the dashed line in Fig. 2) similar to that observed for holes

in Ge.[12] Using the optical-phonon threshold of 0.037 eV[18] and $m_+ = m$ one predicts the optical-phonon-limited positron velocity in Ge to be 5.5×10^6 cm sec^{-1}. Because of the expected simplicity of the positron band, measurements of v_+ at lower temperatures or higher fields than used here might actually be used to measure m_+.

Our mobility measurement may be used to estimate the positron thermalization time in Ge. Using the formalism of Ref. 18 we find that the time t_{12} to slow a positron from energy ϵ_1 to energy ϵ_2 by energy loss to acoustic phonons is

$$t_{12} = \frac{3\pi^{1/2}}{16} \frac{\mu (kT)^{3/2}}{v_l^2} (\epsilon_2^{-1/2} - \epsilon_1^{-1/2}).$$

Taking $\epsilon_1 \gg \epsilon_2 = \frac{3}{2}kT$ we find the thermalization time $\tau = (3\pi/128)^{1/2}(\mu kT / v_l^2) = 1$ psec at 93 K which is short compared to the 2γ annihilation lifetime $(\tau_+ = 220$ psec).[9] Extrapolating by $(kT)^{-1/2}$ implies that positrons should thermalize in Ge at 4 K within ~ 5 psec which is again short compared to τ_+ and consistent with the observations of Ref. 16.

Although the sample used in this experiment was a semiconductor detector, a simple modification of the present experimental arrangement should allow the measurement of μ_+ in many insulators and semiconductors.

Note added.—A further investigation of the Doppler shift versus bias voltage at LN_2 temperature yielded the following data [$V_1 - V_2$, velocity]: [1100 V, $(0.26 \pm 0.09) \times 10^6$ cm sec^{-1}], [1400 V, $(0.43 \pm 0.09) \times 10^6$ cm sec^{-1}], [1650 V, $(0.31 \pm 0.15) \times 10^6$ cm sec^{-1}], consistent with the fitted line in Fig. 2.

[1]D. G. Costello, D. E. Groce, D. F. Herring, and J. W. McGowan, Phys. Rev. B 5, 1433 (1972).

[2]A. Seeger, J. Phys. F 3, 248 (1973).

[3]S. DeBenedetti, C. E. Cowan, W. R. Konneker, and H. Primakoff, Phys. Rev. 77, 205 (1950); R. L. Garwin, Phys. Rev. 91, 1571 (1953); A. Perkins and J. P. Carbotte, Phys. Rev. B 1, 101 (1970).

[4]B. Bergersen and T. McMullen, Solid State Commun. 9, 1865 (1971); R. N. West, Adv. Phys. 22, 263 (1973).

[5]G. Lang and S. DeBenedetti, Phys. Rev. 108, 914 (1957).

[6]O. Sueoka and S. Koide, to be published.

[7]I. K. MacKenzie and J. L. Campbell, Nucl. Instrum. Methods 101, 149 (1972).

[8]R. Paulin, R. Ripon, and W. Brandt, Phys. Rev. Lett. 31, 1214 (1973).

[9]A. Gainotti and C. Ghezzi, J. Phys. C 5, 779 (1972).

[10]B. Rozenfeld, A. Baranowski, and K. Jerie, Phys. Lett. 53A, 154 (1975); F. Heinrich and A. Oggenfuss, Phys. Lett. 55A, 441 (1976).

[11]J. Bardeen and W. Shockley, Phys. Rev. 80, 72 (1950).

[12]G. Ottovani, C. Canali, and A. A. Quaranta, IEEE Trans. Nucl. Sci. 22, 192 (1975).

[13]K. Fujiuara, T. Hyodo, Y. Takakusa, and O. Sueoka, J. Phys. Soc. Jpn. 39, 403 (1975).

[14]The detectors were obtained from Princeton Gamma-Tech, Princeton, N. J. 08540.

[15]We understand that these Ge crystals were supplied to Princeton Gamma-Tech by the General Electric Company, Philadelphia, Pennsylvania.

[16]M. A. Shullman, G. M. Beardsley, and S. Berko, Appl. Phys. 5, 367 (1975).

[17]All the data shown in Fig. 2. were obtained with V_2 = 300 V except for the $V_1 - V_2 = 2800$-V LN_2 datum for which V_2 was 200 V. By observing the charge collection of the positron-induced pulses in the small detector it was determined that the electric field reaches the front surface of this detector ("punch through" occurs) when the bias is between 200 and 300 V. The electric field in the region where the positrons are stopping is therefore less than 100 V per 0.5 cm (the detector thickness) when the bias is 300 V.

[18]Paul Norton and H. Levinstein, Phys. Rev. B 6, 478 (1972).

Concentration-Dependent Kohn Effect in Cubic Tungsten Bronzes*

W. A. Kamitakahara, B. N. Harmon, J. G. Taylor, L. Kopp, and H. R. Shanks

Ames Laboratory—ERDA and Department of Physics, Iowa State University, Ames, Iowa 50010

and

J. Rath

Department of Physics, University of Iowa, Iowa City, Iowa 52240

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Inelastic neutron scattering measurements on single crystals of cubic $Na_x WO_3$ reveal a large concentration-dependent Kohn anomaly in the [100] longitudinal acoustic-phonon dispersion curve. The results demonstrate the two-dimensional character of the Fermi surface, support the rigid-band model for $0.56 < x < 0.83$, and conflict with the percolation model which has just recently been proposed to explain the transport properties of the tungsten bronzes.

A pronounced, concentration-dependent Kohn effect has been observed in crystals of the nonstoichiometric compound $Na_x WO_3$ for x values between 0.56 and 0.83. Comparisons of the shape and x-dependent position of the Kohn anomaly with first principles calculations of the $NaWO_3$ band structure and $\vec{q}$-dependent susceptibility $\chi(\vec{q})$ yield two important conclusions: (1) The Fermi surface is characteristic of a two-dimensional metal in a restricted sense, and (2) the rigid-band model is valid for this system. The results suggest that for the investigation of certain wave-vector–dependent properties, $Na_x WO_3$ can be useful as a prototype two-dimensional metal with the advantages (over real two-dimensional metals, the layered compounds[1]) of large, stable, single crystals and an easily variable electron concentration. In addition, the results are in direct conflict with the percolation model which has very recently been used to explain the

intriguing electronic-transport properties of $Na_x WO_3$.[2]

Sodium tungsten bronze, $Na_x WO_3$, has the cubic perovskite structure for $x \geq 0.5$ above about 150°C. At lower temperatures in this concentration range there is a very small tetragonal distortion[3] which we neglect in the analysis of the experiments and the calculations described below. The neutron scattering measurments were performed on triple-axis spectrometers at the Ames Laboratory Research Reactor. The crystals, grown by fused-salt electrolysis[4] at the Ames Laboratory, were typically cubic in shape, with volumes of approximately 10 cm³. The [100] longitudinal acoustic-phonon dispersion curve was measured for three x values, determined from x-ray diffraction measurements of the lattice constant to be 0.565 ± 0.003, 0.593 ± 0.002, and 0.830 ± 0.007. The majority of the data were taken at a fixed incident energy of 12.9 THz us-