to 6.1 for the LCY simulations and 6.5 for the ST2 calculation. This slightly smaller value and the deeper minimum between the intermolecular peaks imply that the near-neighbor shell of molecules is more tightly coordinated than the simulations predict. Other physically plausible ways of extrapolating between 0 and 1.4 Å^{-1} in Eq. (5) result in smaller coordination numbers and deeper minima than the straight-line approximation. A similar picture emerges from an analysis of the O-H distinct structure factor data, to be reported elsewhere. These yield a hydrogen coordination number of 1.7 about oxygen in good agreement with the LCY simulations.

In conclusion, using the high epithermal flux of a pulsed neutron source and a novel isotope-substitution technique, we have obtained the first model-independent distinct structure factors in liquid water. The results are in good agreement with simulations, but they suggest that the short-range order is more tightly coordinated than predicted by available theories. This method should also be applicable to many other systems with light atoms where inelasticity corrections are significant.

This work was performed under the auspices of the U. S. Department of Energy, with partial support from the Division of Materials Sciences/ Basic Energy Sciences.

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Channeling Radiation from Relativistic Positrons in LiF

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(Received 1 June 1982)

Unusual features of radiation spectra from relativistic positrons channeled in the ionic crystal LiF have been observed.

PACS numbers: 61.80.Mk, 41.70.+t, 71.60.+z, 78.70.-g

We have observed channeling radiation from a binary crystal, in this case from relativistic positrons channeled in the ionic crystal LiF. Some of the features of this channeling radiation are unusual and interesting because of the presence and periodicity of two different ionic species of widely different atomic number (Li, $Z = 3$, and F, $Z = 9$). Along the planes whose Miller indices are all odd (or along axes whose Miller indices sum to an even number) the Li and F ions are aligned separately, so that rather than having planes (or axes) along which the average ionic charge is 6, one has planes (or axes) along which the ionic charge alternates between 3 and 9. The shapes of the interplanar or interstring potentials for these special planes [(111), (113), etc.] and axes [⟨110⟩, ⟨112⟩, ⟨130⟩, etc.] are strikingly different from the relatively simple shapes of the potentials associated with the planes and axes of single-element crystals.

In single-element crystals, the interplanar potentials are approximately harmonic, and the positron channeling-radiation spectrum is nearly monochromatic because of the nearly equal spacing of energy levels.[1] However, the more complicated interplanar potentials for planes such as (111) in LiF should engender a richer spectrum of energy levels for positrons and hence a richer channeling-radiation spectrum. We have observed such a spectrum from positrons channeled along the (111) planes in LiF, and have studied its tilt-angle dependence.

Figure 1 shows the major interplanar potentials that we have calculated for LiF. The eigenfunctions and energy levels were computed with the many-beam approximation, described by Andersen, Eriksen, and Lægsgaard,[2] which takes into account the periodicity of the crystal potential. The Bloch-wave nature of the eigenfunctions becomes apparent only near the tops of the potential wells where the energy levels broaden into bands. The Fourier coefficients U_n of the potentials were derived from the electron scattering factors $f_{el}(s)$ for isolated Li^+ and F^- ions given in Ref. 3. Although these $f_{el}(s)$ for individual ions diverge as $s \rightarrow 0$, the overall charge neutrality of the LiF unit cell ensures that the divergences cancel, so that U_0 is finite. For large values of s, the values of Ref. 3 were extrapolated by adding two terms: (a) the scattering factor for a bare charge, and (b) a Lorentzian which was adjusted for the best fit to each scattering factor. To account for the thermal vibrations of the ions, the scattering factors were multiplied by appropriate Debye-Waller factors. We used the one-dimensional mean-square vibration amplitudes given by Witte and Wolfel[4]: $\langle U_{Li}{}^2\rangle = 0.0139$ Å^2 and $\langle U_F{}^2\rangle = 0.0085$ Å^2. One sees at a glance that our calculated (111) planar potential for positrons [Fig. 1(c)] is a kind of double (nearly harmonic) well within a larger (nearly harmonic) well, much richer in spectral lines than the simple, nearly harmonic wells associated with the (110) and (100) planes [Figs. 1(a) and 1(b), respectively].

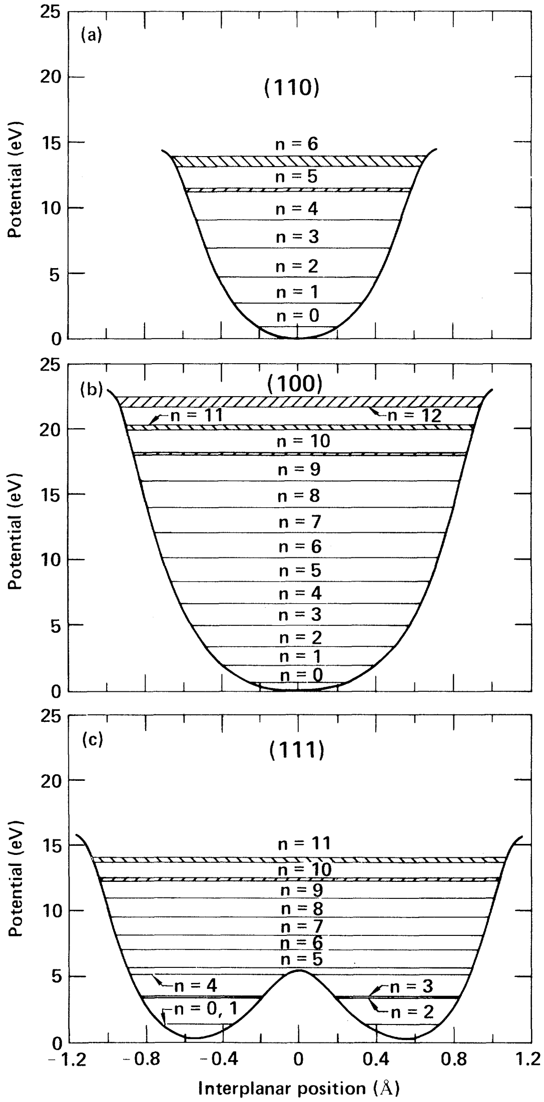

FIG. 1. Calculated interplanar potentials for LiF: (a) for the (110) plane, (b) for the (100) plane, (c) for the (111) plane.

The measurements were carried out with a 54.5-MeV ($\gamma = 107.6$) positron beam ($\Delta E/E = 0.3\%$) of divergence < 0.3 mrad from the Lawrence Livermore National Laboratory electron-positron linear accelerator incident upon LiF crystals (etched with water) having thicknesses of 25 and 125 μm. Most of the details of the experimental apparatus and procedures are given by Berman and co-workers.[5] For the present measurements, the collimators limiting the positron and photon divergences were 0.5 cm in diameter (half the previous values); the former was particularly important in enabling the considerable improvement in beam tuning to be made. Also, a larger (20-cm-diam) plastic scintillator in the swept-beam hole served as the positron beam monitor, making relative normalization more accurate.

The channeling-radiation spectra for the 25-μm LiF crystal are shown in Fig. 2, together with the theoretical predictions based upon the potentials shown in Fig. 1. The good agreement of the results of this relatively crude calculation with the data, especially for the more complex (111) planar case [Fig. 2(c)], demonstrates the essential correctness of this approach, although it can be seen that the calculation tends to yield peak energies for all three planar directions that are somewhat higher than the experimental values. Minor improvements in the theory could be made by inclusion of effects which broaden the higher-lying states (and perhaps decrease their relative intensity as well). Accounting for the distortion of the isolated-ion potentials within the crystal (see Ref. 4) probably would help, as would a more accurate treatment of thermal vibrations.

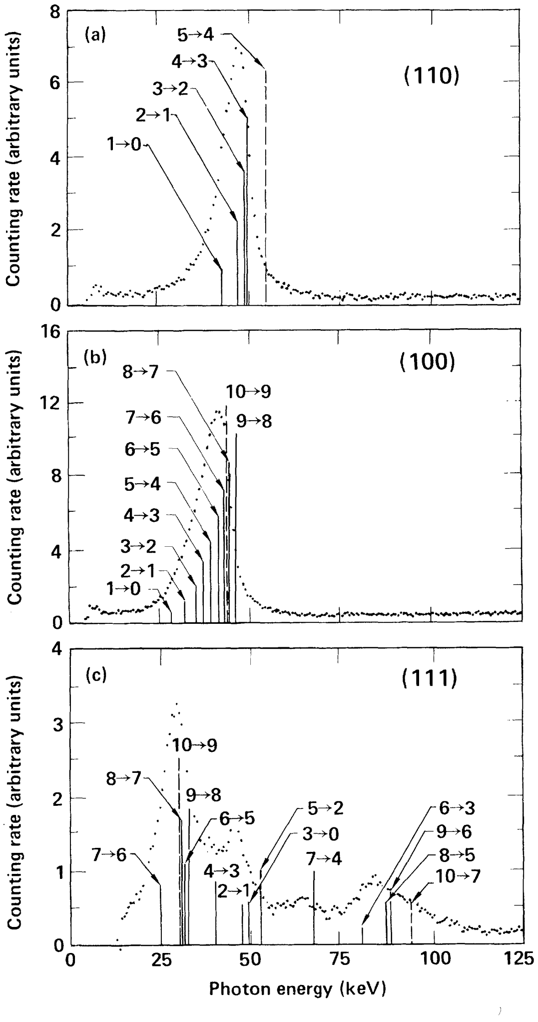

FIG. 2. Measured background-corrected radiation spectra from 54-MeV positrons channeled in LiF: (a) along the (110) plane, (b) along the (100) plane, (c) along the (111) plane. The relative intensities of the spectral lines calculated with the potentials of Fig. 1, assuming equal initial level populations, are shown as vertical lines. The dashed lines are somewhat uncertain because of the proximity of the initial level to the top of the well and its consequent band broadening.

Figure 3 shows the (100) channeling-radiation spectrum over a wider energy range. The peak in the spectrum at 120 keV results from $\Delta n = 3$ transitions between bound states in the potential well of Fig. 1(b), as we have seen before for electrons channeled in silicon.[6] The intensity of these transitions is proportional to the anharmonicity of the potential[7]; for a harmonic potential the matrix elements vanish.

The dechanneling length for 54.5-MeV positrons along the (100) plane in LiF can be estimated from a comparison of the channeling-radiation intensities for crystals of different thicknesses. The measured ratios of the counting rates for channeling radiation and for bremsstrahlung (as approximated from placing the LiF crystal in a random orientation with respect to the incident beam direction), both integrated over the photon energy range from 24 to 57 keV, were 6.2, 4.0, and 3.7 for crystal thicknesses of 25, 125, and 150 μm, respectively. We thus infer that the half-length for decay of the bound-state population is approximately 150 μm.

Finally, the initial population of levels depends

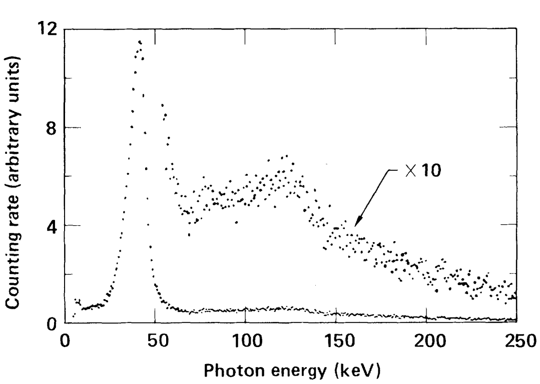

FIG. 3. Measured background-corrected radiation spectrum from 54-MeV positrons channeled along the (100) plane in LiF, showing the $\Delta n = 3$ transitions.

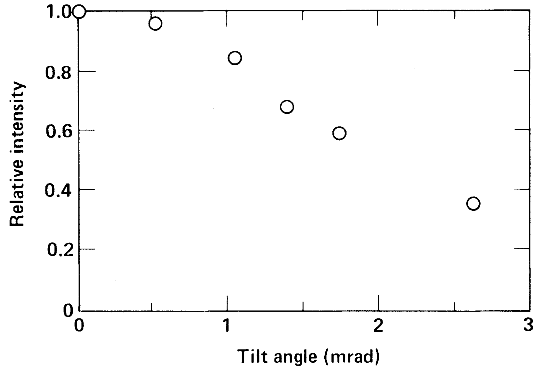

FIG. 4. Measured intensity ratio of the 45 and 30 keV peaks in the radiation spectrum from 54-MeV positrons channeled along the (111) plane in LiF as a function of the tilt angle between the channeling plane and the incident positron beam direction. No error bars are shown because the dominant uncertainties are systematic rather than statistical (see text).

upon the angle between the channeling plane and the incident beam direction (the "tilt angle"), because the positrons have a larger effective transverse energy for larger tilt angles. Therefore, it is reasonable to suppose that with increasing tilt angle, the levels near the top of a potential well would be populated preferentially to the levels that are more tightly bound. The LiF crystal provides us with a laboratory in which to study this effect, since for positrons channeled along the (111) plane, the $n=2$ to 4 levels lie in the "little well" [see Fig. 1(c)] whereas the $n=6$ to 9 levels lie in the "big well." Therefore, the intensities of the $2\to1$, $3\to0$, and $4\to3$ spectral lines which make up the 45-keV peak in Fig. 2(c) should decrease more rapidly with tilt angle than $6\to5$, $7\to6$, $8\to7$, and $9\to8$ lines which make up the 30-keV peak. The measured intensity ratio of the 45- and 30-keV peaks as a function of tilt angle is shown in Fig. 4. Here the regions of integration are from 42.1 to 50.8 and from 23.2 to 38.7 keV, and the entire area under the spectral peaks (including both bremsstrahlung and free-to-bound transition strength) has been stripped off before integration. This stripping process introduces a systematic uncertainty (of the order of 10%) which substantially exceeds the statistical uncertainties. Clearly, this ratio decreases sharply with tilt angle, as expected.

In sum, we have observed the fragmentation of a *positron*-induced channeling radiation spectrum in the (111) planar direction of the ionic crystal LiF, in good agreement with a simple calculation. In addition, $\Delta n = 3$ transitions were observed, the dechanneling length was estimated from the thickness dependence of the channeling-radiation intensity, and the expected tilt-angle dependence of the relative intensities of spectral lines in the "little" and "big" potential wells was observed.

The work was performed at Lawrence Livermore National Laboratory under the auspices of the U. S. Department of Energy under Contract No. W-7405-ENG-48, and was supported in part by the Air Force Office of Scientific Research under Grant No. AFOSR 81-0029 and by the Joint Services Electronics Program under Grant No. DAAG 29-81-K-0057. The authors thank R. Klein for help in the processing of the tilt-angle data.

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