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Separation of Elastically and Inelastically Scattered γ-Radiation from $LiNbO_3$ by Mössbauer Diffraction

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Abstract

Mössbauer diffraction experiments have been performed on a $LiNbO_3$ single crystal at room temperature for scattering angles between 14·5 and 40·5°. A large contribution of inelastically scattered intensity was detectable for all scattering angles. Radiation damage caused by the high-energy components of the radiation emitted by the ^{57}Co Mössbauer source was observed. An attempt was made to estimate the temperature factor B from the measured inelastically scattered radiation assuming that first-order thermal diffuse scattering (TDS) is dominant. Both the TDS contribution to the integrated intensity within the Bragg peak and the ratio of the integrated first-order TDS included in the Bragg peak to that included in the background were calculated with various programs from the elastic constants and compared with the experimental values.

I. Introduction

Ferroelectrics like $LiNbO_3$ are of great interest for technical as well as scientific reasons. A comprehensive review of the properties of $LiNbO_3$ is given by Räuber (1978). Whereas extensive X-ray investigations (Abrahams, Reddy & Bernstein, 1966; Fujimoto, 1982), neutron scattering investigations (Chowdhury, Peckham & Saunderson, 1978) and some Mössbauer experiments on ^{57}Fe-doped crystals (Lauer, Pfannes & Keune, 1979; Pfannes, Lauer, Keune, Maeda & Sakai, 1980; Date, Gonser & Keune, 1979; Date, Joag, Engelmann, Keune & Gonser, 1981) have been reported, to our knowledge no measurements have been performed by means of Mössbauer diffraction. With this method the extraordinary high energy resolution of the Mössbauer effect can be used to measure both the elastically and the inelastically scattered radiation from crystals not containing Mössbauer isotopes. Therefore, it is possible to determine both the contribution of thermal diffuse scattering (TDS) to the intensities of Bragg reflections and the temperature factor B as demonstrated for a silicon single crystal (Krec & Steiner, 1984).

A direct determination of the temperature factor B from ordinary X-ray diffraction experiments is not possible (Lehmann, 1980). Abrahams *et al.* (1966) determined a mean value for B of 0.496 Å^2 from measurements of 247 reflections. Fujimoto (1982) obtained for a single crystal parallel to the hexagonal Z direction, the polar axis of $LiNbO_3$, a B value of 0·385 Å^2. Since these values are not in good agreement and also, an experimental proof of the applicability of TDS correction programs for such rather complicated structure types is desirable, Mössbauer diffraction experiments were performed on a single crystal of $LiNbO_3$ cut perpendicular to the piezoelectric Y axis to obtain additional information on the lattice dynamics.

In § II a short description of the experimental setup and the crystal orientation is given. The results for

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the elastically and inelastically scattered intensities are discussed in § III.

II. Experimental details

A special two-circle goniometer was constructed to obtain a rigid connection between the Mössbauer source [^{57}Co *Rh*, 100 mCi (1 Ci = 37 GBq)] and the resonant absorber (^{57}Fe *Rh*) which was always fixed in position. Therefore, the same area of the absorber was always illuminated and no extra determination of the photoabsorption was necessary. The background relevant for the 14·4 keV ($\lambda = 0{\cdot}8602$ Å) energy interval could be separated because of the energy-dispersive mode of registration and a very precise determination of both the inelastically and the elastically scattered intensities was possible (Fig. 1). A detailed description of the experimental setup is given in the preceeding paper (Krec & Steiner, 1984). All measurements have been performed at room temperature in symmetric Bragg geometry. The diffractometer was driven in θ–2θ scan.

The room-temperature structure of $LiNbO_3$ (space group $R3c$) was determined by Abrahams *et al.* (1966).

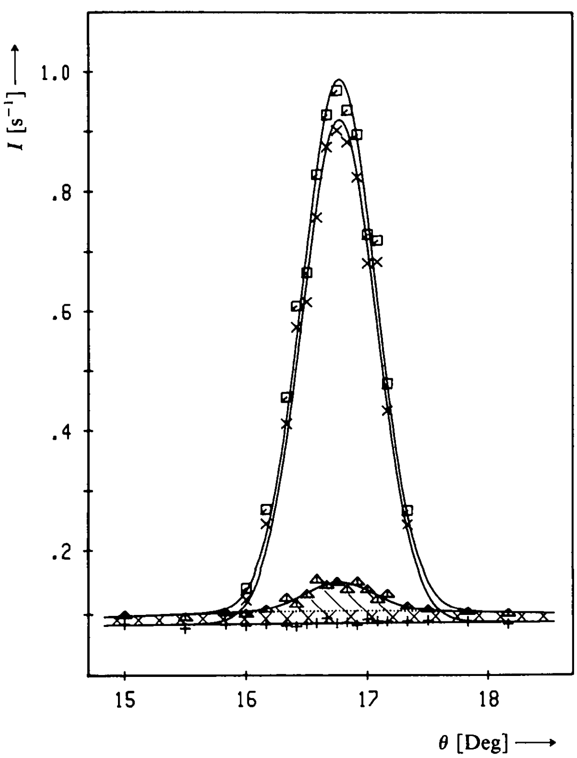

Fig. 1. $LiNbO_3$ 030 reflection. Inelastic (△), elastic (×) and total (□) intensity. Full curves calculated by means of Gaussian distribution functions. Background (+), full curve: linear regression. I^{tot}_{TDS} (hatched and cross-hatched area) total integrated TDS intensity, I^{B}_{TDS} (cross-hatched area) integrated TDS intensity included in the background.

A single crystal of $LiNbO_3$ (supplied by Crystal–Tec*) was cut perpendicular to the piezoelectric Y axis. The surface, containing the hexagonal X–Z plane, was Syton polished. Care was taken to prepare a single-domain plane-parallel plate with overall dimensions of 20.0 × 15.0 × 0.54 mm. The crystal quality was checked by X-ray topography (Willibald, 1981; Pongratz, Cerva, Skalicky, Oppolzer & Willibald, 1982) and the dislocation density was found to be less than 300 mm^{-2}.

III. Results and discussion

A. *Elastically scattered intensity*

Although the investigations started with a nearly perfect single crystal, a distortion of the illuminated surface was observed after the irradiation period necessary to perform the measurements (approximately 4 months for each reflection). The area of damage was greater than the area irradiated by the 14.4 keV radiation. Since the high-energy components of the radiation emitted by the Mössbauer source (122 and 136 keV) were not completely absorbed by the aluminium collimator, only these components were able to produce the observed larger damaged region. The degree of distortion was equally distributed about the whole damaged area. Therefore, the observed radiation damage must be mainly caused by the high-energy components of the source radiation. The surface fraction of the distorted area was estimated in a scanning microscope† to be about 20%. This radiation damage is also indicated by the measured elastic intensities of the two reflections (030 and 060), the line shapes of which were approximated by Gaussian distribution functions (Figs. 1 and 2). Whereas the measured integrated elastic intensity of the 030 reflection was slightly higher than predicted by the dynamical theory, for the 060 reflection an increase in intensity of about 60% was obtained and the intensities of both reflections differ remarkably from those predicted by the kinematical theory (Table 1). All quantities were calculated with a measured linear absorption coefficient of 84·88 cm^{-1}, which is in good agreement with the theoretical one (86·097 cm^{-1}). The atomic scattering factors were taken from *International Tables for X-ray Crystallography* (1974) and the dispersion correction for the niobium atoms was interpolated from the values given by Saravia & Ellis (1966) for a wavelength of 0.8602 Å. The temperature factor B was determined from the separately

* Crystal Technology Inc., 1035 East Meadow Circle, Paolo Alto, California 94303, USA.

† The electron micrograph has been deposited with the British Library Lending Division as Supplementary Publication No. SUP 39278 (2 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

Table 1. *Comparison of measured and calculated elastically scattered integrated intensities* (s^{-1}) *for the two investigated reflections of* $LiNbO_3$

Reflection	Measured (s^{-1})	Calculated (s^{-1}) Dynamical	Kinematical
0 3 0	0·636 (9)	0·608	6·653
0 6 0	0·141 (2)	0·087	0·463

measured inelastically scattered radiation, as described below. Owing to the change in crystal quality caused by radiation damage a determination of *B* from the elastically scattered radiation was not possible. To avoid this damage a double-crystal spectrometer should be used for ferroelectric crystals like $LiNbO_3$.

B. Inelastically scattered intensity

Independent of the elastically scattered intensity which, as discussed above, is strongly influenced by the radiation damage, the inelastically scattered intensity can be measured especially below and in the vicinity of the Bragg peaks. Owing to the energy resolution of the Mössbauer effect radiation with energy changes greater than 10 neV is detected as being inelastically scattered. Within the interval from 14·5 to 40·5° this inelastically scattered intensity was always detectable (Fig. 3) and peaked at the Bragg angle. Elastically scattered contributions were only measured in an angular range in the vicinity of the Bragg peaks (Figs. 1, 2). Owing to the higher Bragg angle the fraction of the inelastic intensity below the Bragg peaks is larger for the 060 reflection ($I_{inel}/I_{el} = 0{\cdot}204$) than for the 030 reflection ($I_{inel}/I_{el} = 0{\cdot}107$). However, when comparing these values the influence of the radiation damage on the measured intensity of the elastically scattered radiation must be considered. The shape of the inelastically scattered intensity below the Bragg peaks is in agreement with that proposed by the TDS theory. Therefore, for the following it was assumed that in the vicinity of the Bragg peak the inelastic intensity remaining after the subtraction of the background (see Fig. 1) was caused by TDS only. Measurements at different temperatures should be performed to confirm this assumption (Schneider, 1980).

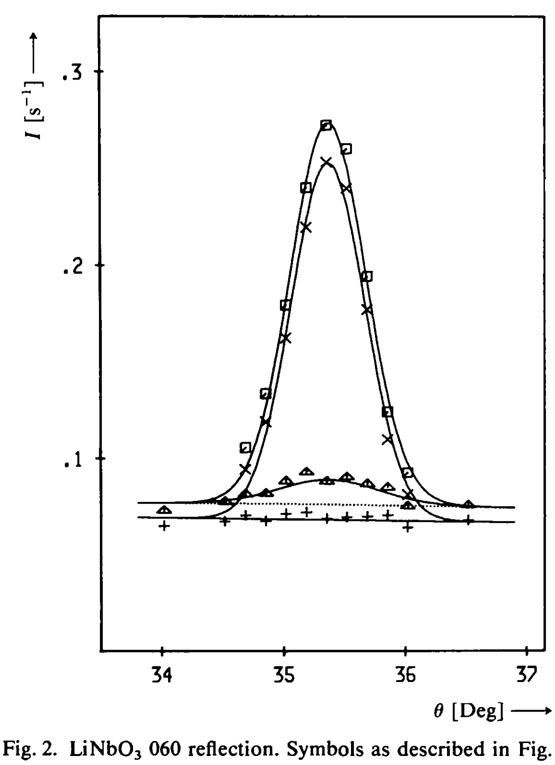

Fig. 2. $LiNbO_3$ 060 reflection. Symbols as described in Fig. 1.

As observed for a silicon single crystal (Krec & Steiner, 1984) the agreement of the measured and the calculated ratio of the total first-order TDS intensity included in the Bragg peak (I^{tot}_{TDS}) to the one included in the background (I^{B}_{TDS}) was rather poor (Table 2) if the illuminated volume was approximated by a sphere or a cylinder and the anisotropy of the elasticity was neglected (Willis, 1969). By means of several computer programs based on the long-wavelength continuum elasticity approximation (Stevens, 1974; Kurittu & Merisalo, 1977; Helmholdt & Vos,1977) these two TDS contributions were also calculated from the elastic constants reported by Smith & Welsh (1971). The ratios of $I^{B}_{TDS}/I^{tot}_{TDS}$ are in complete agreement and differ from the measured ones by 10 and 12% for the 030 and 060 reflections, respectively (Table 2). Since, owing to the radiation damage, the elastically scattered intensity could not be determined and to avoid errors resulting from uncertainties in the determination of the primary intensity, only the ratios I^{tot}_{TDS} (060)/I^{tot}_{TDS} (030) [0·401, 0·400 and 0·400 after Stevens (1974), Helmholdt & Vos (1977) and Kurittu & Merisalo (1977), respectively] should be compared with the measured one (0·414). It follows that for a scan length of 2° the measured ratio of the

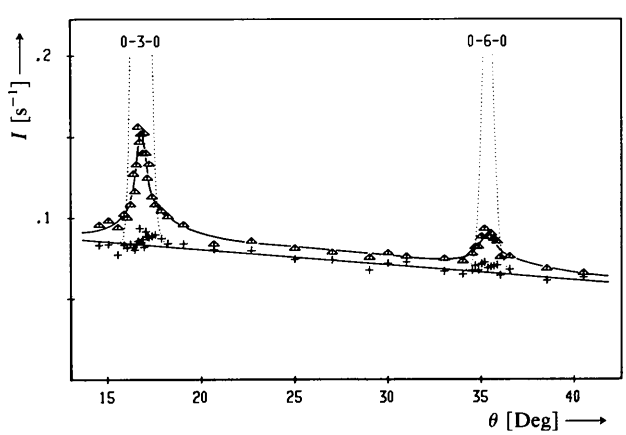

Fig. 3. Inelastic intensity for $LiNbO_3$, symmetric Bragg case, θ–2θ scan. Symbols as described in Fig. 1.

Table 2. *Measured and calculated ratios* I^B_{TDS}/I^{tot}_{TDS}

I^B_{TDS} is the integrated first-order TDS intensity included in the background, I^{tot}_{TDS} is the integrated first-order TDS intensity included in the Bragg peak. Scan length 2·0°.

		Calculated[a] approximation		Numerical calculations		
Reflection	Measured	cylinder	sphere	St[b]	H & V[c]	K & M[d]
030	0·52 (7)	0·15	0·33	0·574	0·577	0·577
060	0·56 (9)	0·32	0·33	0·497	0·498	0·498

Calculated after: (*a*) Willis (1969); (*b*) Stevens (1974); (*c*) Helmholdt & Vos (1977); (*d*) Kurittu & Merisalo (1977).

TDS contribution can be taken into account by these first-order TDS calculations with an accuracy of approximately 4%.

Assuming that (i) the harmonic approximation is valid, (ii) the influence of the radiation damage on the diffuse intensities is negligible and (iii) the first-order TDS is still the most dominant contribution, a mean temperature factor B can be determined from the measured inelastic intensities of at least two reflections with parallel scattering vectors **S** (for details see Krec & Steiner, 1984). The measured inelastically scattered radiation included in the Bragg peak was integrated after subtracting the contribution in the background. The resulting integrated intensity was corrected for second-order TDS contributions, the amount of which was estimated with the program *TDS*2 written by Stevens (1974). From the ratio of the resulting diffuse intensities of the two reflections B was determined using the known structure factors F_o. The obtained B values at 295 K are 0·37 (8) and 0·33 (8) $Å^2$ for the cylindrical and spherical approximations used for the calculation of the illuminated volume and are in agreement with the one reported by Fujimoto (1982) measured in the hexagonal Z direction.

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Signal, Noise and Resolving Power in Rotation Searches

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Abstract

First and second moments of the probability density of the function $R=\sum_H E_o^2E_c^2$ are evaluated for n-atom models consisting of i correctly and $n-i$ incorrectly placed atoms of an N-atom structure. Formulas are valid for space groups $P1$ and $P\bar{1}$, and describe the influence of the size of the model as well as data truncation. Introduction of the concept of an averaged structure leads to structure-independent conclusions about the behaviour of the resolving

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