Determination of anomalous scattering factors in GaAs using x-ray refraction through a prism

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The advantages of synchrotron radiation have been employed to revitalize a 60-year-old technique for measuring anomalous scattering factors by accurately measuring x-ray refraction through a prism. We report results obtained from a GaAs sample in the vicinity of both the Ga and As K absorption edges. Our analysis of the technique shows that relative accuracies of 0.03 electrons and absolute accuracies of 0.1 electrons should be readily obtainable. This should be adequate for the needs of anomalous x-ray scattering measurements.

I. INTRODUCTION

Considerable interest is emerging in anomalous x-ray scattering as a useful technique to investigate local order in both solids and liquids, due principally to the advent of synchrotron radiation (SR) sources with the capability of producing intense x-ray beams at arbitrary energies close to a large number of atomic K and L absorption edges. The basic technique proceeds analogously to the technique of isotopic substitution in neutron scattering experiments. That is, scattering from an ensemble is linearly decomposed into scattering from a set of atomic partial distribution functions, each weighted by a pair of atomic scattering factors. A series of measurements made at energies which significantly change the atomic scattering factors can then be inverted to obtain the partial distribution functions, provided the changes in atomic scattering factors are known to sufficient accuracy.[1] In principle, these partial distribution functions describe the constituent atoms' average local environments species by species out to distances of about 10 Å. However, both theoretical and experimental analyses indicate that the anomalous factors, f', must be absolutely accurate to approximately 0.1 electron if the technique is to succeed.[2,3] Thus, in practice to date, only differential distribution functions have been accurately determined[3,4,5] since f' accuracies of only 0.3 to 0.5 electrons appear to be required. While these differential distribution functions give only the species averaged local environments of the constituent atoms, they have still proven to be an exceptionally powerful tool for understanding the structure of concentrated noncrystalline materials, including liquids.[3,5]

Unfortunately, for experimentalists there are few literature values of f' available which are accurate to either level and fewer still near to absorption edges. Recently some results have been obtained by x-ray interferometry,[6,7] but the technique is laborious and has so far been limited to a few pure metals. The need for accurate f' values on samples of choice has thus stimulated a search for new measurement techniques. These have included Bragg intensity measurements,[8] refraction measurements through a wedge using a monolithic Laue-Laue diffractometer,[9] and conversion of absorption cross sections to f' by use of the Kramers-Kronig dispersion relation.[10] Theoretical determinations of f' using calculated atomic wave functions[11,12] are only accurate far from absorption edges, becoming unreliable in their immediate vicinity, particularly in the presence of white lines or strong chemical effects in multicomponent systems. To date, none of these approaches has satisfied the needs for both accuracy and ease of application which will be required of a generally viable technique. This then was the context in which we undertook the revival of x-ray refractive index measurements in prisms in order to assess their potential as an f' determination technique.

II. DESCRIPTION OF TECHNIQUE

A. Principle

The basic geometry of the technique is indicated in Fig. 1(a). An incoming x-ray beam impinges at angle θ upon the face of a prism. Entering the new medium it is refracted, making angle γ to the backside of the face. It leaves the prism at nearly normal incidence, so the refraction at this interface is negligible, and has a final angle of deviation β from the direction of the incoming beam, where the indicated angles satisfy $\beta+\gamma=\theta$. The angle θ will typically be only slightly larger than the critical angle θ_c for the prism material and therefore β will be relatively large, of the order of θ_c itself. If we represent the real part of the index of refraction of the medium in the standard notation as $1-\delta$, neglect refraction at the exit surface, and assume δ for the external medium is negligible, then Snell's law operates only once and may be written

$$(1-\delta)\cos\gamma=\cos\theta \tag{1}$$

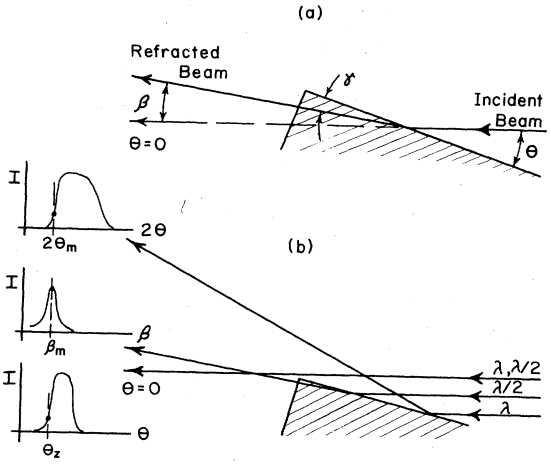

FIG. 1. Refraction measurement through the corner of a prism. (a) The basic concept. (b) As implemented using a SR beam with harmonics.

or, in the small angle approximation,

$$\theta^2-\gamma^2=2\delta=\theta_c^2 \tag{2}$$

in which

$$\delta=(\lambda^2e^2/2\pi mc^2)\sum_a(Z_a+f_a')N_a\ , \tag{3}$$

where all the terms have their usual meanings and N_a is the number of type a atoms per unit volume. In Eq. (2) we have noted explicitly the relation between δ and the critical angle θ_c.[13] Thus if the angle θ is known, a measurement of β produces a value of δ.

This technique was first applied by Larsson, Siegbahn, and Waller in 1924 (Ref. 14) and then employed by Larsson,[15] Stauss,[16] and Bearden[17] in the following decade. James has a good summary of these efforts.[18] For the most part, the technique was used to measure the ratio e/m or as a calibration technique for the wavelength λ. To our knowledge, only Larsson actually used this method to measure f' close to an absorption edge (Ca in calcite).[15]

Our hope is to be able to measure f' to 0.1 electrons. A substitution of typical parameter values into Eq. (2) shows that angular resolutions of about 5 μrad are required for both θ and β to obtain f' absolutely to this accuracy. If only relative f' changes are required, only β must be so accurately determined. The nature of the SR source assists us here, since, for entrance slits of 50 μ or less, the input beam divergence is approximately 15-μrad full width at half maximum (FWHM), arising entirely from the 300-μ FWHM source size at 20 m. In the absence of complicating factors, it should be straightforward to determine centroid shifts of 5—10 % of this value, obtaining accuracies of 0.03 electrons or better. Hence we set up our experiment with the geometry shown in Fig. 1(b). We note that the incoming SR beam from a precisely tuned two-crystal Bragg monochromator has a high harmonic content. Hence we tune for wavelength λ, where $\lambda/2$ corresponds to our energy of interest, and set θ to satisfy the relationship $\theta_c(\lambda/2)<\theta<\theta_c(\lambda)$. Thus λ is totally externally reflected, while $\lambda/2$ is refracted. If the beam is adjusted vertically so part passes over the prism, then this provides a $\theta=0$ reference marker. To measure $f'(E)$, the input wavelength is scanned over the energy region of interest while θ remains fixed. θ is typically about 0.3° and β is about 0.15°. The diffractometer 2θ arm has a resolution of 4000 steps/degree, which is adequate for our desired precision.

B. Relative merits

The most important merit of this technique is that the angles to be measured scale as θ_c, which in turn is proportional to the square root of δ. Since typical values of δ are in the 10^{-4} to 10^{-8} range, the prism technique gains a factor of 10^2 to 10^4 advantage over techniques where deflections are directly proportional to δ. This includes both the wedge measurement mentioned above[9] and measurements of the influence of refraction on Bragg angle, where $n\lambda=2d\sin\theta(1-\delta/\sin^2\theta)$.[19]

A second advantage is that the prism remains fixed during the experiment, with only the input energy being changed. Hence the stability of the beam path, the illuminated portion of prism, and the angle of incidence are all easy to maintain. As $\lambda/2$ is changed only the relative motion of the angle β must be measured. Thus the technique should be particularly good for measuring relative changes in δ.

A third advantage is that only the positions of the three beams are important. There is no need to measure either relative or absolute intensities.

The major technical problem associated with the technique is that the edge of the prism must be of very high quality. A calculation of absorption lengths at glancing angle shows that only about the top 1000 Å of the prism are actually sampled. Hence the prism must be square, without edge rounding to at least an order of magnitude better than this (i.e., less than 100 Å). This has so far restricted investigations to cleavable crystals. However, we believe that modern lithographic and reactive ion etching techniques should be applicable here, making it possible to cut an appropriate step a few microns deep in any material whose surface can be prepared for total external reflection of x rays.[20] In particular, the technique could then be employed on vapor-deposited materials, which can often be made only in the form of very thin films.

III. EXPERIMENTAL CONDITIONS

The experiments were done on the materials diffractometer located on the IV-3 wiggler line at the Stanford Synchrotron Radiation Laboratory. The input monochromator crystals were Si(220), giving 0.2-eV resolution at $\lambda/2$ equal to 11 KeV. Input slits of 7.5 μ were located 0.5 m upstream of the prism. This slit size gave good beam definition and also reduced extraneous air scatter into the detector, since less than a micron of beam can actually be refracted. Additional Al filters were inserted into the line to measure the position of the direct and externally reflected beams. The exit slits were set to 25 μ

at 0.5 m from the sample. The detectors were scintillation counters, which had adequate energy resolution to reject harmonics. A sample of GaAs was cleaved from a 2-in. wafer provided by the Center for Materials Research at Stanford, and examined in a scanning electron microscope at 20000×. At this enlargement the edge was perfect over its length and no rounding could be observed.

IV. EXPERIMENTAL RESULTS

A. Determination of incident angle θ

The results of a typical diffractometer scan are shown in Fig. 2. The direct beam at angle θ_z is seen to be extremely sharp. The refracted beam, at angle β, is about 0.04° FWHM and regularly shaped with a slightly sloping background. The reflected beam has a sharply rising leading edge, followed by a rich peak structure, which we will discuss later. Since the positions and shapes of both direct and reflected beams were found to be essentially energy independent, they were measured only infrequently and only the refracted beam region was scanned as a function of energy.

The value of 2θ is energy independent for a given prism orientation and is the angle between the small angle sides of the direct and reflected beam profiles [see Fig. 1(b)]. We modeled these edge profiles as arising from a plane wave cut by "Fresnel edge," for which the true edge position lies at the point where the intensity reaches one-quarter of its mean jump.[21] This worked well for the direct beam. The unexpected oscillations in the reflected beam above the edge produce some uncertainty in exactly what the mean jump is, but the edge itself is sufficiently steep so that reasonable results can still be obtained. As we shall see, the major effect of θ errors is to produce offsets in the resultant f' values, but not to affect relative accuracy. These errors are examined in detail in Sec. V.

B. Determination of refracted angle β

Figure 3 shows five typical refracted beam profiles, and shows how the peak position and shape change with energy as we move across the Ga absorption edge. Two effects are present. First, as we cross the edge, f' decreases to a minimum and then increases again. This corresponds to the peaks' shift first to smaller β and then back again with increasing energy. Second, as the edge is crossed, photoelectric absorption rises dramatically, greatly reducing the transmission of the refracted beam through the prism. This both reduces and broadens the peaks, the latter being a diffraction effect accompaning the decrease in height ("source size") of the beam at it exits the prism.

Two sets of data were collected. The first steps through the Ga edge in increments of about 1 eV at an incidence angle of 0.298°. The second is a much sparser set through the As edge at $\theta=0.223°$. θ was decreased here to accommodate the decrease in θ_c with increasing energy. Peak positions were determined by removing backgrounds and computing the centroid of that portion of each peak which was greater than 25% of its maximum. Curves of refracted angle position in the two energy ranges are shown in Figs. 4(a) and 4(b). Notice that the absolute angles are about 0.18° and show changes of about 0.02°.

C. Determination of f' values

Several steps are required to convert peak positions into f' values. First, peak positions are converted into β values by subtracting 0.0635°, the direct beam position marking the true diffractometer zero. β and θ values are then substituted into Eq. (2) to obtain $\delta(E)$. The δ values found here were in the range of 8×10^{-6} near the Ga edge and 6×10^{-6} near the As edge. Equation (3) can then be solved for the sum $f'_{\mathrm{Ga}}+f'_{\mathrm{As}}$, assuming N_{Ga} and N_{As} are known. N_{Ga} equals N_{As} and both may be obtained from the known crystal structure and density of GaAs. The resultant f' sums near the two edges are shown in Figs. 5(a) and 5(b), respectively. For purposes of comparison, these two figures also show f' sums computed using the theory of Cromer and Liberman (CL),[11] which is known

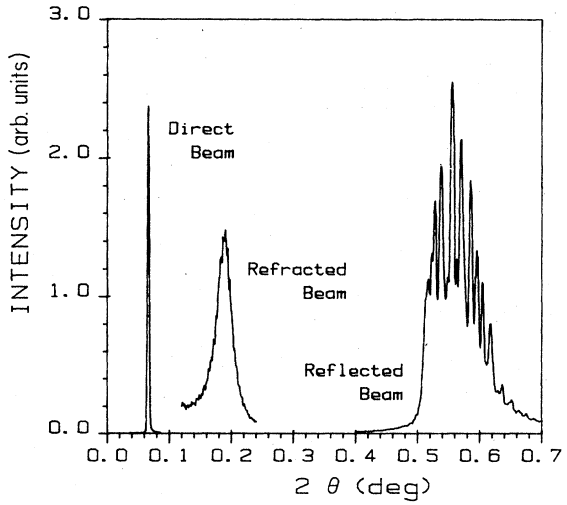

FIG. 2. Diffractometer scan through the three beams indicated in Fig. 1(b).

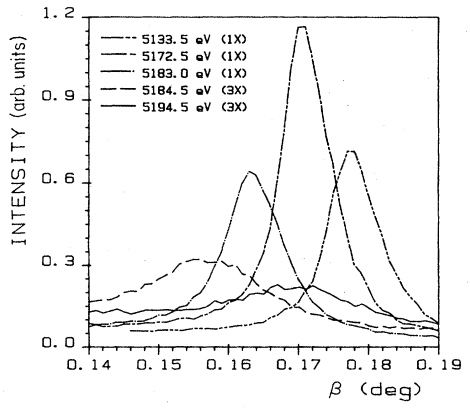

FIG. 3. The refracted beam at five typical energies, showing centroid shifts and shape changes.

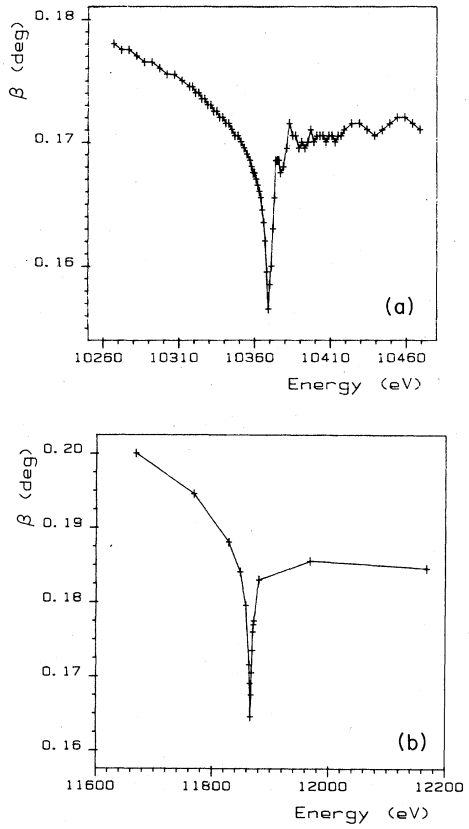

FIG. 4. Refracted beam centroid positions versus energy for a GaAs prism. (a) Near the Ga edge. (b) Near the As edge.

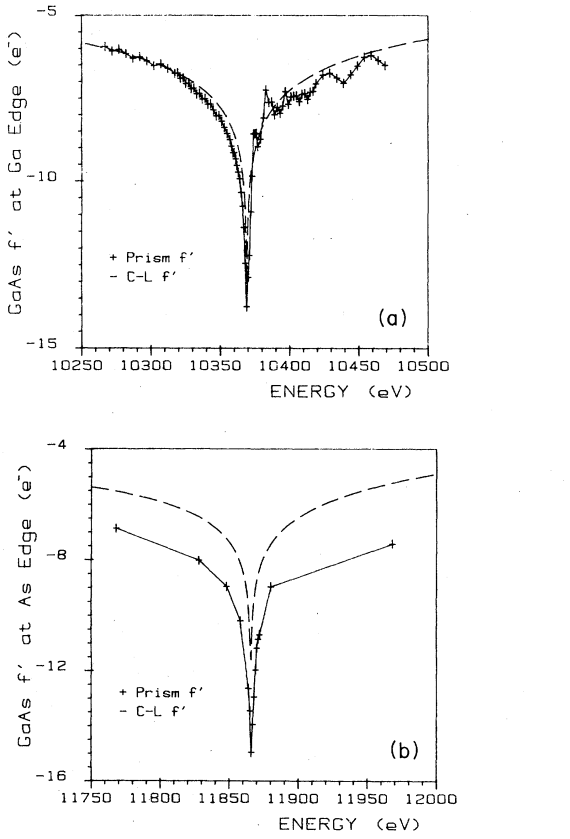

FIG. 5. Anomalous scattering factor f' values versus energy. Shown are values measured in the GaAs prism (+) and calculated using Cromer-Liberman theory. (a) Near the Ga edge. (b) Near the As edge.

to be relatively accurate well below absorption edges. The Ga edge data appear to agree remarkably well with the CL theory at low energies, while the As edge data disagree by about 1.3 electrons.

As a further aid to discussion of our results, we present them again in Fig. 6, this time accompanied by two additional curves. The first is the imaginary anomalous scattering factor f'', obtained by application of the optical theorem to extended x-ray absorption fine structure (EXAFS) data on GaAs.[4,10] The second is an estimate of f' obtained by integration of the Kramers-Kronig (KK) integral relating f' and f''. (See Ref. 10 for details of the technique. Instead of using McMaster f'' values to evaluate the integral outside the experimental range as in Ref. 10, we used CL f'' values which are usually more accurate. The discrepancy between the two approaches is smaller than 0.2 electrons.)

V. DISCUSSION AND ANALYSIS

A. Features of the curves

The qualitative features of f' determined by the prism technique are extremely encouraging. The expected features all occur: a smoothly decreasing curve below the edge, an extremely sharp spike at the edge corresponding to absorption white lines, and fine structure above the edge, which is equivalent to EXAFS absorption fine structure. Scatter in the data below the edge lead to a rms error estimate of 0.1 electrons. Above the edge it is unclear whether lower peak heights have led to more scatter in the data or whether the measurement is faithfully reproducing the EXAFS fluctuations, but with a higher resolution than in the EXAFS-derived results. Further study will be necessary, but three factors support the latter conclusion. The first is the presence of the sharp spike slightly above the edge in the Ga prism f' data which corresponds to the presence of the second Ga white line in the EXAFS data. This shows only as a shoulder in the KK-derived f' curve. Secondly, the fractional change in f' at the minimum is significantly larger in the prism data. Finally, the fluctuations in the EXAFS region do not occur point to point, as they would in a truly random situation, but have periods of the same order and minima and maxima in the general positions as in the KK-derived data.

The major quantitative difference between the prism f' and KK f' data is the large offset of about 1.3 electrons observed below the As edge. This is clearly an error in the prism data, since the KK data agree fairly well with CL computed values at energies far from the edge, where the theoretical calculations are fairly accurate. In addition, the offset between the prism data and either the KK or CL estimates is systematically about 0.5 electrons larger

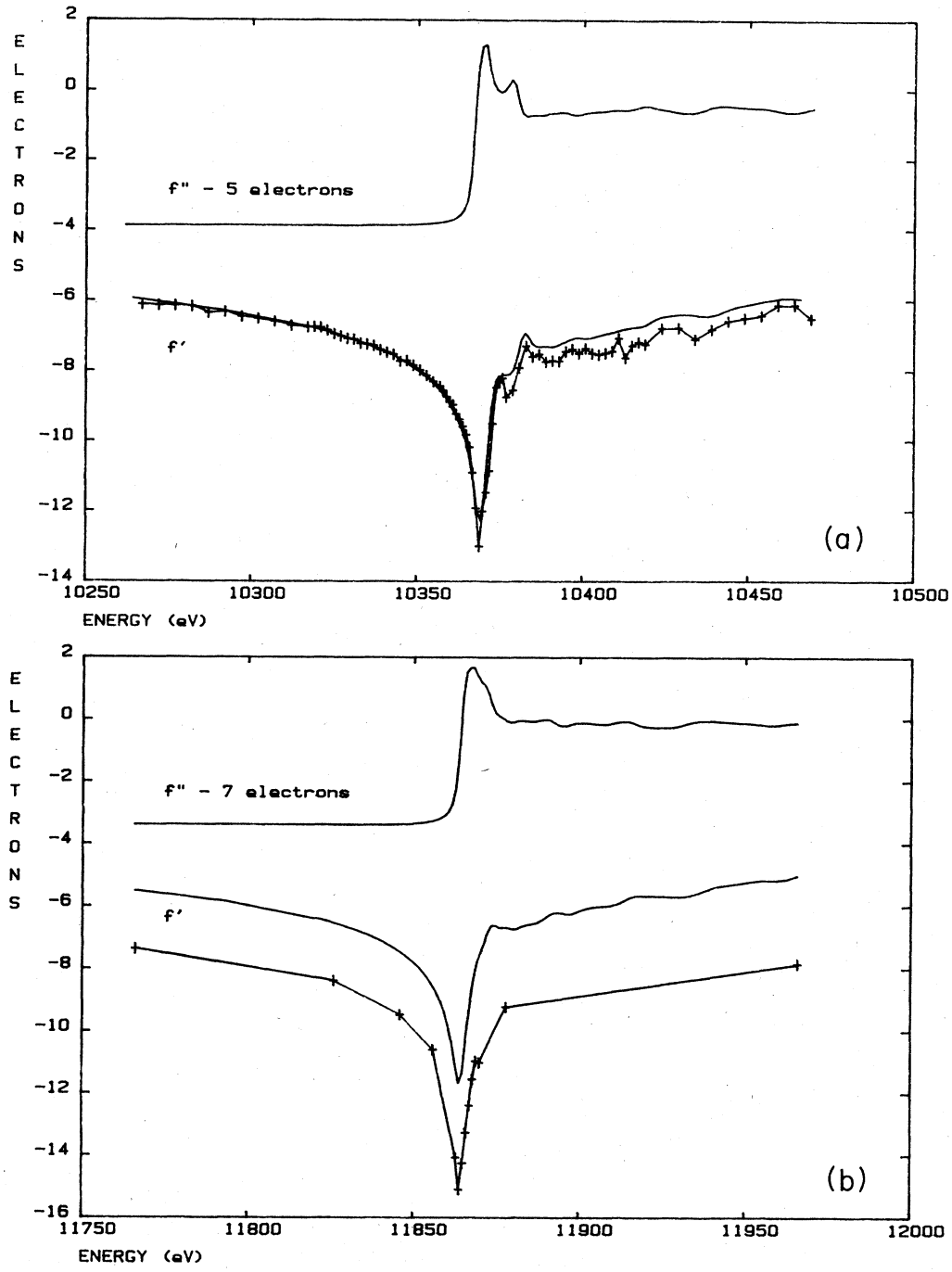

FIG. 6. A comparison between f' values obtained from the prism measurement and those obtained by Kramers-Kronig transformation of f'' data from an EXAFS absorption measurement. (a) Near the Ga edge. (b) Near the As edge.

above the edges than below. This differential offset is more likely to be significant than the absolute offset, which is easily caused by errors in the measurement of θ. We are currently investigating the modification of Snell's law due to absorption as a possible explanation for both of these discrepancies.

B. Error estimation

In order to estimate our errors it is necessary to solve Eqs. (2) and (3) for f' and expand them to show the explicit dependence on measured quantities. These are θ_m, the measured value of θ; β_m, the measured angle of refraction; and θ_z, the measured zero angle. They are related to θ and β by $\theta=\theta_m-\theta_z/2$ and $\beta=\beta_m-\theta_z$. This gives

$$f'=\frac{mc^2}{e^2}\frac{\pi}{N\lambda^2}(2\theta_m\beta_m-2\theta_m\theta_z-\beta_m^2+\beta_m\theta_z)$$
$$-(Z_a+Z_b)\,, \tag{4}$$

from which partial derivatives with respect to the three variables may be easily obtained. These are

$$\frac{\partial f'}{\partial \theta_m} = C(\beta_m - \theta_z), \quad \frac{\partial f'}{\partial \beta_m} = C(\theta_m - \beta_m + \theta_z/2),$$
$$\frac{\partial f'}{\partial \theta_z} = -C(\theta_m - \beta_m/2), \tag{5}$$

where $C=(2mc^2\pi)/(e^2N\lambda^2)$, and is approximately $7.0E+6$ at the Ga edge. For example, at 61 eV below the Ga edge, we have $\theta_m=0.3300°$ $(+0.0016°/-0.0008°)$; $\theta_z=0.0635°$ $(+0.0010°/-0.0005°)$; and $\beta_m=0.174°$ $\pm 0.0003°$. Generous error estimates were obtained for the θ measurements by noting the change in θ required to move to three-fourths and one-eighth of the Fresnel edge jump, respectively. The error estimate for β_m is also generously estimated at 5% of the refracted beam FWHM. Conversion to radians and substitution into Eq. (5) gives estimates of the independent contributions to f' error as

$$\frac{\partial f'}{\partial \theta_m} = +1.4E4\ e^-/\text{rad}, \quad \Delta f'|_{\theta_m} = +0.38/-0.19 \text{ electrons}, \tag{6a}$$

$$\frac{\partial f'}{\partial \beta_m} = +2.3E4\ e^-/\text{rad}, \quad \Delta f'|_{\beta_m} = \pm 0.12 \text{ electrons}, \tag{6b}$$

$$\frac{\partial f'}{\partial \theta_z} = -3.0E4\ e^-/\text{rad}, \quad \Delta f'|_{\theta_z} = +0.52/-0.26 \text{ electrons}. \tag{6c}$$

Adding the larger errors in quadrature gives a mean expected error of 0.65 electrons, which agrees fairly well with the observed difference between our data and the KK f' values. In addition, the estimate in Eq. (6b) accounts quite nicely for the observed scatter in the data below the absorption edge where we expect a smooth curve. This analysis demonstrates that the absolute accuracy depends most strongly on the accurate determination of θ_z and θ_m and reflects the difficulty in obtaining an accurate edge location. We note that with some effort it is reasonable to expect to reduce β_m errors by a factor of 5, to 1% of the peak FWHM, giving errors of 0.03 electrons. To obtain similar errors from the θ measurements, however, increases of accuracy of 13 and 17 are needed for θ_m and θ_z, respectively, suggesting that considerable modification of the technique for determining these angles may be necessary. Without such modifications the current precision can probably only be increased by a factor of about 2, for an absolute accuracy of about 0.3 electrons.

We therefore see that precision is more readily obtained from the prism measurement than is absolute accuracy, given our current apparatus. This suggests that even without further refinements the technique could be a convenient means for measuring the deviations in f' from Cromer-Lieberman values which arise in the vicinity of absorption edges due to chemical or structural effects. This would be done by adjusting θ_m and θ_z in Eq. (4) to match f' to the CL values far from the edge, where they are known to be accurate, and then using the measurement of β_m to determine f' closer to the edge. We found, in the case of the As edge data, that a shift in θ_z of less than 0.05° was needed to superimpose the two curves at 11 765 eV. Changes in either θ_z or θ_m produced essentially uniform offsets, but in opposite directions [see Eqs. (6)]. The two effects are so strongly anticorrelated that only adjustments in a single parameter are appropriate in the suggested procedure.

C. Technical improvements

The first improvement needed is in the diffractometer 2θ arm. If we wish to determine f' to 0.03 electrons, Eq. (6b) shows that we need precision to about 1 μrad with a range of less than 1°. This is a heavy demand to place on even a very-well-made gear tooth. Instead, the exit slits should be mounted on a precision slide and driven with a precision micrometer. An 0.5-mm/turn micrometer, directly driven by a 1000 step/revolution gear motor has just this angular resolution and accuracy.

Some improvement in our technique for determining θ_z and θ_m would also be useful. If improving our ability to take finer θ steps is insufficient to obtain the desired result, then perhaps some modification of the Laue-Laue diffractometer employed by Deutsch and Hart[9] would be useful.

Finally, there is the question of the structure in the reflected beam, which is complicating our determination of θ_m. We have considered, as its source, the following possibilities: diffraction from our entrance slit, interference in an oxide film on the GaAs surface, and surface figure undulations. The shape of the reflected beam profile does strongly suggest some interference phenomena. The profile is, for example, strongly reminiscent of the interference pattern generated by a pair of slits illuminated by a partially coherent source.[22] The synchrotron source is indeed partially coherent, but we have only a single slit and the pattern observed in this case is essentially dominated by the central diffraction lobe.[23] The telling argument against this hypothesis, however, is that no similar structure is observed in the direct beam. Using a model similar to ones employed for describing x-ray reflection from layered synthetic multilayers,[24] we investigated the possibility of interference in a surface oxide layer. A curve similar to the reflected beam could be produced, but an oxide layer of 1000 Å was required to do so. This can

be ruled out, based on the experience of the semiconductor industry that native oxides on GaAs are very thin, typically only 2 to 3 atomic layers thick.[25] Furthermore, since our measurement depth is of the same order, the fact that we obtain essentially correct results using N_a computed from the density of pure GaAs discards this hypothesis. On the other hand, even surfaces prepared as x-ray reflector show local undulations in surface slope which produce distortions in a reflected beam of the type we have observed.[23] Commericaly polished Si crystals are typically even worse.[26] Even with a 7.5-μ input slit, at θ equals 0.298° and 0.223°, 1.4 and 1.9 mm of the GaAs surface are sampled, respectively, so there is latitude for such undulations to occur. In future experiments we will test these possibilities more carefully by making an accurate study of the direct beam with no sample inserted, by testing additional materials whose surface oxide state is well characterized, and by testing the surface reflectivity in the presence and absence of the prism corner. If surface figure is indeed found to be a problem then either the samples will have to be prepared more carefully, for example by deposition on x-ray reflection flats, or we shall have to develop methods for determining the incidence angle θ without employing the total external reflection technique.

VI. CONCLUSIONS

Through the application of SR techniques we have been able to measure the refraction of x rays through a prism of GaAs with sufficient accuracy to determine the anomalous scattering factor f' near both Ga and As K absorption edges. The technique has excellent precision, which we estimate could be readily extended to 0.03 electrons and its energy resolution is limited only by the resolution of the input monochromator. The absolute accuracy is about 0.7 electrons and still needs improvement, principally in determining the angle of incidence and the zero of 2θ. With improvement in this area, the technique offers promise as an easy way (given a SR source) to obtain absolute f' values for use in scattering measurements on relatively arbitrary solid samples using essentially the same equipment as for the scattering measurement itself. Even without further improvement, the technique is adequate for measuring deviations in f' from theoretical Cromer-Lieberman values due to chemical effects near the absorption edge.

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