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Diffuse Scattering from Defect Clusters near Bragg Reflections*

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The diffuse scattering from crystals containing defect clusters with strong displacement fields has been calculated. The scattering is extremely large and concentrated in small regions near the Bragg reflections. For small concentrations the diffuse intensity is essentially determined by a "cluster form factor" depending explicitly on the displacement field. The scattering is studied in detail for small and large deviations from the Bragg reflection. Exact expressions are given for the moments of the scattering, depending sensitively on the displacement field in the core of the cluster. The diffuse scattering for strong displacement fields shows a pronounced asymmetry; its center does not coincide with the position of the Bragg peak, in agreement with recent experimental observations.

I. INTRODUCTION

The diffuse scattering from crystals containing defects with displacement fields was first considered by Eckstein[1] and Huang.[2] This theory has been worked out in more detail by Cochran and Kartha[3,4] and by Borie,[5] and has also been the subject of a recent paper of Keating[6] which has generated some controversy.[7] A number of excellent papers about diffuse scattering of defects have been published by Krivoglaz and co-workers,[8,9] but they seem to have been widely overlooked.

The diffuse scattering from defect clusters has also been considered by Krivoglaz and Ryaboshapka,[10] who calculated the scattering from dislocation loops. According to them, the diffuse scattering from loops is extremely large near the Bragg reflections and, under certain conditions, the center of the diffuse scattering can be displaced from the position of the Bragg peak.

Recently the diffuse scattering from defect clusters has been observed experimentally.[11–16] The present author's observation of the diffuse scattering of dislocation loops in neutron-irradiated Cu crystals has been the reason to consider the scattering from defect clusters in somewhat more detail.

II. GENERAL FORMULAS FOR THE DIFFUSE SCATTERING

In this section we derive the general formulas for the elastic scattering from lattices with defects. In the following we are mainly interested in defects with strong displacement fields. Such large displacements are typical for defect clusters rather than isolated point defects; e.g., for a dislocation loop the characteristic displacement is a Burgers vector or a lattice constant. The method we use is essentially that of Krivoglaz[8,9] except that we consider the image field explicitly. For simplicity we take only the scattering at the displaced matrix atoms into account, since this is the only important contribution near the Bragg reflection. The diffuse intensity is given by the scattering function:

$$S(\vec{K}) = \sum_{m,m'} e^{i\vec{K}\cdot(\vec{R}^m-\vec{R}^{m'})} \times \left(\langle e^{i\vec{K}\cdot(\vec{u}^m-\vec{u}^{m'})}\rangle - \langle e^{i\vec{K}\cdot\vec{u}^m}\rangle \langle e^{-i\vec{K}\cdot\vec{u}^{m'}}\rangle \right) . \quad (1)$$

Here $\vec{K}=\vec{k}-\vec{k}'$ is the scattering vector, $\vec{R}^m$ the position of atom m in the *ideal* lattice, and $\vec{u}^m$ the static displacement from this position. The first term in the bracket gives the total intensity, and we have subtracted the Bragg intensity (second term) to obtain the diffuse intensity alone.

The displacement $\vec{u}^m$ is a superposition of the displacements $\vec{t}^{\,m,n}$ due to defects at the different positions n. We introduce a random number $s^n = 1$ or 0, if the position n is occupied by a defect or not (e.g., impurity, vacancy or center of defect cluster, dislocation loop, etc.),

$$\vec{u}^m = \sum_n s^n \vec{t}^{\,m,n} . \quad (2)$$

Further, we assume that the different defects are not correlated, i.e., the numbers s^n and $s^{n'}$ are independently distributed. Then the averages in (1) can be calculated using the relations $(s^n)^2 = s^n$

and $\langle s^n \rangle = c$, where c is the concentration of defects per lattice site. For example,

$$\langle e^{i\vec{K}\cdot\vec{u}^m} \rangle = \Pi_n \langle e^{is^n \vec{K}\cdot\vec{t}^{m,n}} \rangle = \Pi_n [1 + c(e^{i\vec{K}\cdot\vec{t}^{m,n}} - 1)]$$

$$= \exp \sum_n \ln [1 + c(e^{i\vec{K}\cdot\vec{t}^{m,n}} - 1)]$$

$$\simeq \exp c \sum_n (e^{i\vec{K}\cdot\vec{t}^{m,n}} - 1) \ . \qquad (3)$$

For the last step we have assumed that $c \ll 1$, which is normally the case. However, the following arguments hold as well for the exact expression [with $\ln(1+x)$ instead of x]. Thus we have

$$S(\vec{K}) = \sum_{m,m'} e^{i\vec{K}\cdot(\vec{R}^m - \vec{R}^{m'})} \times [\exp c \sum_n (e^{i\vec{K}\cdot(\vec{t}^{m,n} - \vec{t}^{m',n})} - 1)$$

$$- \exp c \sum_n (e^{i\vec{K}\cdot\vec{t}^{m,n}} + e^{-i\vec{K}\cdot\vec{t}^{m',n}} - 2)] \ . \qquad (4)$$

Now $\vec{t}^{m,n}$ consists of two contributions. The first, a displacement $\vec{t}^{m-n}$ due to the "real forces," is the exact solution in an infinite crystal. $\vec{t}^{m-n}$ is highly localized near $m = n$, depends only on $\vec{R}^m - \vec{R}^n$, and falls off as $|\vec{R}^m - \vec{R}^n|^{-2}$ for large distances. The second contribution $\vec{\tau}^{m,n}$ is due to the "image forces" being introduced to satisfy the boundary condition for a finite crystal. Characteristically, $\vec{\tau}^{m,n}$ is slowly varying over the crystal dimensions and very small. The order of magnitude of this term is $\sim \Delta V / R_{\text{cryst}}^2$, where ΔV is the volume change per defect and R_{cryst} a typical crystal dimension.

Now in Eqs. (3) and (4) the displacements $\vec{t}^{m,n}$ are very small for large $|\vec{R}^m - \vec{R}^n|$, and we have in this case [as well as for the exact "ln" in Eq. (3)]

$$c \sum_n (e^{i\vec{K}\cdot\vec{t}^{m,n}} - 1)$$

$$\rightarrow ic \sum_n \vec{K}\cdot(\vec{t}^{m-n} + \vec{\tau}^{m,n}) \sim O(c R_{\text{cryst}}/a) \gg 1. \qquad (5)$$

Here the symbol $O(\ldots)$ means order of magnitude of $\ldots$. Due to the summation over the whole crystal this term can be very large compared with one even for relatively small c, because the average displacement $\langle \vec{u}^m \rangle = c \sum_n \vec{t}^{m,n}$ depends explicitly on the crystal dimensions and is macroscopic, i.e., much larger than a lattice constant. It diverges for $R_{\text{cryst}} \rightarrow \infty$. Therefore even for small concentrations an expansion of $S(\vec{K})$ in the form of Eq. (3) is not possible. This has been overlooked in Ref. 6, leading to the conclusion that the diffuse scattering should be peaked at the unexpanded Bragg reflections (see also Ref. 7). Therefore we now introduce the position $\langle \vec{R}^m \rangle$ in the expanded "average" lattice and get from (4)

$$S(\vec{K}) = \sum_{m,m'} e^{i\vec{K}\cdot(\langle\vec{R}^m\rangle - \langle\vec{R}^{m'}\rangle)}$$

$$\times \{\exp c \sum_n [e^{i\vec{K}\cdot(\vec{t}^{m,n} - \vec{t}^{m',n})} - 1 - i\vec{K}\cdot(\vec{t}^{m,n} - \vec{t}^{m',n})]$$

$$- \exp c \sum_n [e^{i\vec{K}\cdot\vec{t}^{m,n}} + e^{-i\vec{K}\cdot\vec{t}^{m',n}} - 2 - i\vec{K}\cdot(\vec{t}^{m,n} - \vec{t}^{m',n})]\}, \qquad (6)$$

with

$$\langle \vec{R}^m \rangle = \vec{R}^m + \vec{u}^m = \vec{R}^m + c \sum_n (\vec{t}^{m,n} + \vec{\tau}^{m,n}) \ .$$

When compared with Eq. (4) this formula has the advantage that in the exponents we can neglect the image field $\vec{\tau}^{m,n}$ (but not in $\langle \vec{R}^m \rangle$!). By expanding the exponents in powers of $\vec{\tau}^{m,n}$, the linear term of $\vec{\tau}^{m,n}$ alone cancels, and all other terms are extremely small, e.g., the order of magnitude of the sum is

$$\sum_n (e^{i\vec{K}\cdot\vec{t}^{m,n}} - 1) i\vec{K}\cdot\vec{\tau}^{m,n}$$

$$\sim \sum_n \vec{K}\cdot\vec{t}^{m-n}\vec{K}\cdot\vec{\tau}^{m,n}$$

$$\sim O[(V/V_c)^2 a/R_{\text{cryst}}] \ll 1 \ . \qquad (7)$$

The same result holds for $\sum_n (\vec{K}\cdot\vec{\tau}^{m,n})^2$, etc. Therefore, in the exponents inside the bracket $\{\}$ we can go over to a very large (infinite) crystal, and only the "real field" $\vec{t}^{m-n}$ remains. This is the justification of Krivoglaz's treatment,[9] which starts with the lattice homogeneously expanded by the "image" forces and then takes only the "real" displacements into account.

For large distances $|\vec{R}^m - \vec{R}^{m'}|$ the bracket $\{\}$ in (6) goes to zero, which is due to the subtraction of the Bragg scattering in (1). Therefore $S(\vec{K})$ contains no δ functions as the Bragg scattering does.

By introducing the reciprocal lattice vectors $\vec{h}$ of the expanded average lattice ($e^{i\vec{h}\cdot\langle\vec{R}^m\rangle} = 1$) we can always write $\vec{K}$ as $\vec{K} = \vec{h} + \vec{q}$ where $\vec{q}$ lies in the first Brillouin zone. Further, we restrict ourselves to defect clusters with a mean radius R_0 much larger than the lattice constant. Then the displacement field $\vec{t}^{m-n} + \vec{t}(\vec{R}^m - \vec{R}^n)$ is slowly varying from atom to atom and can be calculated using continuum theory. In (6) the summation over n can be replaced by integration. For the same reason we get appreciable intensities only for small values of $q \sim O(1/R_0)$, i.e., in the forward direction ($\vec{h} = 0$) or near Bragg reflections ($\vec{h} \neq 0$). Therefore the summation over m can also be replaced by an integration. If N is the number of lattice sites, we get

$$S(\vec{K}) = S_{\vec{h}}(\vec{q}) = N \int \frac{d\vec{R}}{V_c} \, e^{i\vec{q}\cdot\vec{R}} (e^{\phi(\vec{R})} - e^{\phi(\infty)}) \ , \qquad (8)$$

with

$$\phi(\vec{R}) = c \int \frac{d\vec{r}}{V_c} \{ e^{i(\vec{h}+\vec{q})\cdot[\vec{t}(\vec{R}+\vec{r})-\vec{t}(\vec{r})]} - 1 - i(\vec{h}+\vec{q})\cdot[\vec{t}(\vec{R}+\vec{r})-\vec{t}(\vec{r})] \} \quad (9a)$$

or

$$\phi(\vec{R}) = -2L_{\vec{h}+\vec{q}} + c \int \frac{d\vec{r}}{V_c} (e^{i(\vec{h}+\vec{q})\cdot\vec{t}(\vec{R}+\vec{r})} - 1) \times (e^{-i(\vec{h}+\vec{q})\cdot\vec{t}(\vec{r})} - 1) \quad (9b)$$

and

$$\phi(\infty) = -2L_{\vec{h}+\vec{q}} = -2c \int \frac{d\vec{r}}{V_c} [1 - \cos(\vec{h}+\vec{q})\cdot\vec{t}(\vec{r})] . \quad (10)$$

L is the Debye-Waller factor due to the static displacements which reduce the Bragg scattering.[9,17] For the displacements we have assumed inversion symmetry $\vec{t}(\vec{r}) = -\vec{t}(-\vec{r})$.

For large clusters the differential intensity, as given by Eqs. (8)–(10), is strongly concentrated near the Bragg reflection. Therefore it is difficult to measure the differential intensity directly. However, it is relatively easy to take rocking curves of the diffuse scattering with the use of a double crystal spectrometer as done in Ref. 15. The connection of these curves with the differential intensity is discussed in the Appendix.

III. SMALL CONCENTRATIONS

For small concentrations, i.e., $2L \ll 1$, $S(\vec{q})$ can be expanded linearly in ϕ [Eq. (8)]. In this case the result simplifies because of $\phi(\vec{R})$ being a convolution, the Fourier transform of which is a product:

$$S_{\vec{h}}(\vec{q}) = cN |G_{\vec{h}}(\vec{q})|^2 , \quad (11)$$

with

$$G_{\vec{h}}(\vec{q}) = \int \frac{d\vec{r}}{V_c} e^{i\vec{q}\cdot\vec{r}} [e^{i(\vec{h}+\vec{q})\cdot\vec{t}(\vec{r})} - 1] . \quad (12)$$

Here we want also to give the general result for small c, when the direct scattering at the defects is taken into account, too. For example, we may have interstitials forming interstitial clusters. Let $\vec{R}^I$ be the interstitial position in the unit cell, $\rho(\vec{R})$ the interstitial density within a cluster, and c as above the concentration of clusters per lattice site. Then we have instead of (11)

$$S_{\vec{h}}(\vec{q}) = cN |e^{i\vec{h}\cdot\vec{R}^I} \tilde{\rho}(\vec{q}) + G_{\vec{h}}(\vec{q})|^2 , \quad (13)$$

with

$$\tilde{\rho}(\vec{q}) = \int \frac{d\vec{r}}{V_c} e^{i\vec{q}\cdot\vec{r}} \rho(\vec{r}) .$$

This result permits a simple interpretation: The diffuse scattering is equal to the number of clusters cN times the absolute square of a *cluster form factor*, consisting of two parts. The first, $\tilde{\rho}(\vec{q})$, describes the "direct" scattering at the interstitials in the cluster; the second one, $G_{\vec{h}}(\vec{q})$, describes the "indirect" scattering at the displaced "cloud" of lattice atoms in or near the cluster.

For $\vec{h} = 0$ we get the well-known results for small-angle scattering.[18] Due to the small value of $q \sim 1/R_0$ we get for practically all concentrations

$$S_0(\vec{q}) = cN |\tilde{\rho}(\vec{q}) + G_0(\vec{q})|^2 \quad (14)$$

with

$$G_0(\vec{q}) = \int \frac{d\vec{r}}{V_c} e^{i\vec{q}\cdot\vec{r}} i\vec{q}\cdot\vec{t}(\vec{r}) = -\int \frac{d\vec{r}}{V_c} e^{i\vec{q}\cdot\vec{r}} \partial_{\vec{r}}\cdot\vec{t}(\vec{r}) .$$

The first term in (14) is due to the local density increase produced by the interstitials. The second term, being the Fourier transform of the dilation $-\partial_{\vec{r}}\cdot\vec{t}(\vec{r})$, is due to the local density decrease of the dilated lattice near the cluster. (Because this term partly cancels the first one, the small-angle scattering for clusters with strong displacement fields is typically an order of magnitude smaller than for "normal" clusters.[18])

Near the Bragg reflections, for $\vec{h} \neq 0$, the situation is quite different. First we can replace $e^{i(\vec{h}+\vec{q})\cdot\vec{t}}$ by $e^{i\vec{h}\cdot\vec{t}}$, because $q \sim 1/R_0 \ll h$. Therefore $G_{\vec{h}}(\vec{q})$ now explicitly contains the displacements $\vec{h}\cdot\vec{t}(\vec{r})$, in contrast to $G_0(\vec{q})$ which contains only the dilatation $\partial_{\vec{r}}\cdot\vec{t}(\vec{r})$. Because $\partial_{\vec{r}}\cdot\vec{t}(\vec{r})$ varies appreciably only over a distance R_0, $G_{\vec{h}}(\vec{q})$ is larger than $G_0(\vec{q})$ and $\tilde{\rho}(\vec{q})$ by a factor of about hR_0. {Assuming for Cu[15] a radius of $R_0 = 30$ Å and $\vec{h} = [111]$, the intensities near the Bragg reflection are larger by a factor $(hR_0)^2 \simeq 10^4$ than the small-angle intensities.} Equation (11) is therefore justified so that near the Bragg reflections we "see" essentially the displaced lattice region near the cluster and not the defects themselves as in small-angle scattering. For example, for dislocation loops with radius R_0, the displaced region is essentially a sphere with the volume $\frac{4}{3}\pi R_0^3$ being much larger than the volume $b\pi R_0^2$ of the loop plane.

It is interesting that near the Bragg reflection $S_{\vec{h}}(\vec{q})$ and also $G_{\vec{h}}(\vec{q})$ can be split up into a part $S_{\vec{h}}^S(\vec{q})$ symmetrical with respect to $\vec{q}$ and an antisymmetrical part $S_{\vec{h}}^A(\vec{q})$. From (11) we get

$$S_{\vec{h}}^S(\vec{q}) = S_{\vec{h}}^S(-\vec{q}) = cN\{|G_{\vec{h}}^S(\vec{q})|^2 + |G_{\vec{h}}^A(\vec{q})|^2\} ,$$
$$S_{\vec{h}}^A(\vec{q}) = -S_{\vec{h}}^A(-\vec{q}) = cN 2 G_{\vec{h}}^S(\vec{q})\, G_{\vec{h}}^A(\vec{q}) , \quad (15)$$

with

$$G^S_{\vec{h}}(\vec{q}) = -\int \frac{d\vec{r}}{V_c} \cos(\vec{q}\cdot\vec{r})\,[1-\cos\vec{h}\cdot\vec{t}(\vec{r})]\,,$$
$$G^A_{\vec{h}}(\vec{q}) = -\int \frac{d\vec{r}}{V_c} \sin(\vec{q}\cdot\vec{r})\sin\vec{h}\cdot\vec{t}(\vec{r})\,. \qquad (16)$$

$S^{S,A}$ and $G^{S,A}$ are not only even or odd with respect to $\vec{q}$, but also with respect to the displacement field $\vec{t}$ (and the reflection vector $\vec{h}$). Therefore, for instance, the asymmetry changes if we go over from an interstitial cluster to a vacancy cluster. The asymmetry itself is important only for relatively large displacements (or high-order reflections), for which $\cos\vec{h}\cdot\vec{t}$ and $\sin\vec{h}\cdot\vec{t}$ are comparable, but disappears for $\vec{h}\cdot\vec{t}\ll 1$. Moreover, this asymmetry should not be confused with the size effect[5] representing [in (13)] the interference between $G_{\vec{h}}(\vec{q})$ and $\tilde{\rho}(\vec{q})$ and being neglected in (12).

In order to get a qualitative survey of $S_{\vec{h}}(\vec{q})$ we note that in (12) $(e^{i\vec{h}\cdot\vec{t}(\vec{r})}-1)$ can be replaced by $i\vec{h}\cdot\vec{t}(\vec{r})$ for $r\gg R_{cl}$ whereas for $r\lesssim R_{cl}$ $e^{i\vec{h}\cdot\vec{t}(\vec{r})}$ oscillates more or less rapidly, if $\vec{h}\cdot\vec{t}$ is large as we assume. For a rough approximation, we set therefore for a spherical cluster of n_{cl} point defects with displacement fields $A\vec{r}/r^3$ (cluster model of Ref. 17):

$$e^{i\vec{h}\cdot\vec{t}(\vec{r})} = \begin{cases} iAn_{cl}\,\vec{h}\cdot\vec{r}/r^3 & \text{for } r>R_{cl} \\ 0 & \text{for } r<R_{cl} \end{cases} \qquad (17)$$

and get

$$G_{\vec{h}}(\vec{q}) = -\frac{4\pi A}{V_c}\, n_{cl}\, \frac{\vec{h}\cdot\vec{q}}{q^2}\, \frac{\sin qR_{cl}}{qR_{cl}} - \frac{\frac{4}{3}\pi R_{cl}^3}{V_c}\, 3\, \frac{\sin qR_{cl} - qR_{cl}\cos qR_{cl}}{(qR_{cl})^3}\,. \qquad (18)$$

For $q\ll 1/R_{cl}$, $S_{\vec{h}}(\vec{q})$ is proportional to $1/q^2$ (Huang scattering), whereas for $q\gg 1/R_{cl}$, $S_{\vec{h}}(\vec{q})$ decreases more rapidly, proportional to $1/q^4$. Moreover, the scattering is symmetric for small q, as long as the first term in (18) dominates, and becomes unsymmetric for larger $q\approx 1/R_{cl}$. For $A>0$ (interstitials, etc.), we have more intensity for $\vec{h}\cdot\vec{q}>0$, i.e., for larger Bragg angles.

IV. BEHAVIOR FOR SMALL q VALUES

For very small q values $q\ll 1/R_0$ the diffuse intensity can be evaluated exactly for arbitrary concentrations. From Eq. (8) it follows that the behavior of $S_{\vec{h}}(\vec{q})$ for small q is determined by the behavior of $\phi(\vec{R})$ for large R. According to (9b), $\phi(\vec{R})$ approaches $-2L_{\vec{h}}$ for $R\to\infty$. For large R the deviation of $\phi(\vec{R})$ from $-2L_{\vec{h}}$ can therefore be expanded. It depends only on the asymptotic displacement field $\vec{t}(\vec{r})$ for large $r\gg R_0$. Therefore we get for $S_{\vec{h}}(\vec{q}) = S^S + S^A$:

$$S^S_{\vec{h}}(\vec{q}) = c\,(N/V_c^2)\,e^{-2L_{\vec{h}}}\,|\vec{h}\cdot\tilde{\vec{t}}(\vec{q})|^2 \sim 1/q^2\,, \qquad (19)$$

$$S^A_{\vec{h}}(\vec{q}) = (N/V_c)\,2L_{\vec{h}}\,e^{-2L_{\vec{h}}}\,\vec{h}\cdot\tilde{\vec{t}}(\vec{q}) \sim 1/q\,. \qquad (20)$$

Here $\tilde{\vec{t}}(\vec{q})$ is the Fourier transform of the asymptotic displacement field and is proportional to $1/q$. The symmetric part S^S diverging as $1/q^2$, therefore, dominates in the small-q region (Huang scattering), whereas the antisymmetric part diverges only as $1/q$.

For larger concentrations both terms are reduced by the Debye-Waller factor e^{-2L} just as is the Bragg scattering. Therefore the Huang scattering is unimportant for large concentrations.

As an example we calculate $\tilde{\vec{t}}(\vec{q})$ for a cluster of n_{cl} point defects characterized by three equal crossed dipole moments of strength P_{11}. We have for a cubic crystal

$$\tilde{t}_i(\vec{q}) = \begin{cases} n_{cl}\,\dfrac{P_{11}}{q}\,\dfrac{\kappa_i}{c_{11}}\,, & \text{isotropy} \\ n_{cl}\,\dfrac{P_{11}}{q}\,\dfrac{\kappa_i}{c_{44}+d\kappa_i^2}\left(1+\sum_j \dfrac{c_{44}+c_{12}}{c_{44}+d\kappa_j^2}\,\kappa_j^2\right)^{-1}, & \text{anisotropy} \end{cases} \qquad (21)$$

with

$$\kappa_i = q_i/q, \quad d = c_{11}-c_{12}-2c_{44}\,.$$

In Fig. 1(a) we have for this case plotted the curves of equal intensity of the *symmetrical part* S^S for a $h00$-type reflection using the elastic constants of Cu. The dashed lines represent the curves for elastic isotropy. The full lines are for anisotropy. The intensity vanishes for $\vec{q}\perp\vec{h}$. Also in more general cases, we always have such a "nodal plane," because this is connected with the assumed inversion symmetry $[\tilde{\vec{t}}(\vec{q}) = -\tilde{\vec{t}}(-\vec{q})]$. However, the inclination of this nodal plane will depend on the type of the defects considered. Therefore, if we have several types of defects, we have to sum in (19) over the different contributions. Due to the fact that the different nodal planes will in general not coincide, we get intensity everywhere around the reciprocal lattice point. As an example we consider the scattering from dislocation loops.[17] For a loop with radius R_0 and with Burgers vector $\vec{b}=b\vec{b}^0$ perpendicular to the loop plane πR_0^2 we have in the isotropic case:

$$\tilde{\vec{t}}(\vec{q}) = \frac{b\pi R_0^2}{q}\left(2\vec{b}^0(\vec{\kappa}\cdot\vec{b}^0) + \frac{\nu}{1-\nu}\,\vec{\kappa} - \frac{1}{1-\nu}\,(\vec{b}^0\cdot\vec{\kappa})^2\vec{\kappa}\right) \quad \text{for } q\ll 1/R_0\,. \qquad (22)$$

Assuming, for example, that we have loops statistically distributed on all ⟨111⟩ directions, we get for S^S the curve plotted in Fig. 1(b) $[\vec{h}^0 = h00,\ \nu=\frac{1}{3}]$.

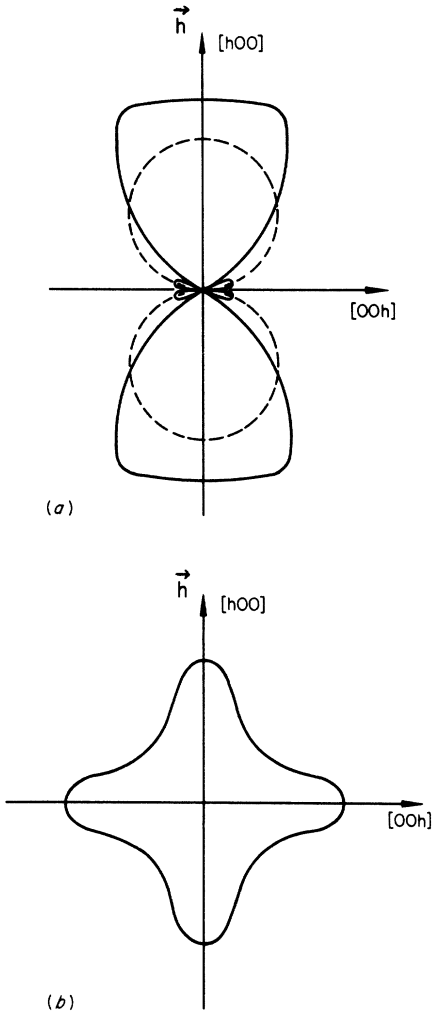

FIG. 1. Curves of equal intensity for the symmetrical part $S^S_{\vec{h}}(\vec{q})$ for a $h00$ reflection in Cu. (a) Cluster of point defects with three equal crossed dipoles; dashed line, isotropy; solid line, anisotropy. (b) Dislocation loops on all {111} planes (isotropy).

For both models (21) and (22)[17] the asymptotic field $\vec{t}(\vec{r})$ is proportional to the number n_{cl} of point defects in the cluster ($n_{cl} = b\pi R_0^2/V_c$ for loops). Therefore, $S^S(\vec{q})$ [Eq. (19)] is proportional to cNn_{cl}^2 for small q, a result which is due to the "coherent" action of all the defects in the same cluster and which is well known in small-angle scattering ($\vec{h}=0$).

The antisymmetric part S^A (20) changes sign at the nodal plane $\vec{h}\cdot\vec{t}(\vec{q})=0$. For a cluster of interstitials (21) we have plotted the curves of constant intensity of S^A in Fig. 2. The ± signs indicate the regions for which $S^A \gtrless 0$, dashed lines again for the isotropic case, full lines for anisotropy. We have more intensity for $\vec{h}\cdot\vec{q}>0$, i.e., apparently larger Bragg angles, which would be reversed for vacancy-type defects. This is just opposite to the change of the Bragg angles of the Bragg intensities due to the lattice expansion.

We get qualitatively the same result for dislocation loops, e.g., randomly distributed on all {111} planes. As a rule, for interstitial-type clusters we always have more intensity for larger Bragg angles, whereas vacancy-type clusters give more intensity for lower Bragg angles (see also Sec. VII).

V. BEHAVIOR FOR LARGE q

Whereas the scattering for small q, i.e., the Huang scattering and the Bragg scattering, is determined by the behavior of $\phi(\vec{R})$ [Eq. (9)] for large $\vec{R}\to\infty$, the scattering for large q values should be determined by the behavior of $\phi(\vec{R})$ for small R values. For this case we have

$$\phi(\vec{R}) = c\int \frac{d\vec{r}}{V_c}\left\{e^{i\partial_{\vec{r}}\cdot[\vec{h}\cdot\vec{t}(\vec{r})]\vec{R}} - i\partial_{\vec{r}}\cdot[\vec{h}\cdot\vec{t}(\vec{r})]\vec{R} - 1\right\} . \tag{23}$$

For large q, small P, respectively, we may expand $e^{\phi(\vec{R})}$ linearly in ϕ. Whereas the term $(1-e^{\phi(\infty)})$, as well as the second terms in (23), give no contribution for very large q values, the first term in (23) gives

$$S_{\vec{h}}(\vec{q}) = cN[(2\pi)^3/V_c^2]\int d\vec{r}\,\delta(\vec{q}-\delta\vec{h}(\vec{r})) , \tag{24}$$

with

$$\delta\vec{h}(\vec{r}) = -\partial_{\vec{r}}(\vec{h}\cdot\vec{t}(\vec{r})).$$

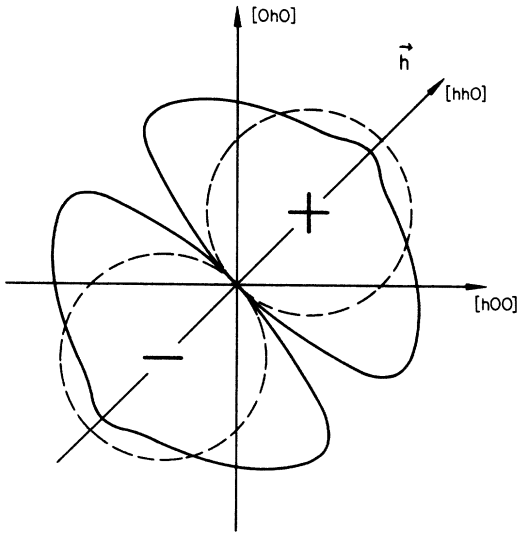

FIG. 2. Curves of equal intensity for the antisymmetrical part $S^A_{\vec{h}}(\vec{q})$; cluster of interstitials with three equal crossed dipoles in Cu, $\vec{h}=hh0$; dashed line, isotropy; solid line, anisotropy.

The quantity $\delta\vec{h}(\vec{r})$, representing the changes of the atomic displacements normal to the diffracting planes, can be interpreted as the "local" change of the reciprocal lattice vector $\vec{h}$. The "macroscopic" change of $\vec{h}$ due to the real field alone is given by c/V_c times the volume integral of $\delta\vec{h}(\vec{r})$. Equation (24) is equivalent to the approximation of Stokes and Wilson,[19,20] being widely used for the scattering from dislocations.[21,22]

If we replace $\vec{t}(\vec{r})$ in (24) by the asymptotic displacement field of the cluster, i.e., $\vec{t}(\vec{r}) \sim n_{cl} A/r^2$ (or $\sim b\pi R_0^2/r^2$ for loops), then we get

$$S_{\vec{h}}(\vec{q}) \sim cN \frac{4\pi A}{V_c^2} n_{cl} \frac{h}{q^4} \quad (\text{or } cN \frac{b\pi R_0^2}{V_c^2} \frac{h}{q^4} \text{ for loops}). \tag{25}$$

Thus, $S_{\vec{h}}(\vec{q})$ decreases as $1/q^4$ and is, contrary to the scattering for small q [(14), (19), (21)] only proportional to n_{cl}.

For the isotropic field $\vec{t}(\vec{r}) = c\,\vec{r}/r^3$, Trinkhaus[20] has evaluated $S_{\vec{h}}(\vec{q})$ for large q starting from Eq. (11) and using the saddle-point method. He gets the same result as obtained in the Stokes-Wilson approximation (24) except for a factor with an oscillating q dependence.

For larger q values $q \gg 1/R_0$ Eq. (25) is no longer valid because $\vec{t}(\vec{r})$ does not increase as $1/r^2$ in the core of the cluster, but more or less levels off. Therefore $S_{\vec{h}}(\vec{q})$ decreases more rapidly than $1/q^4$. For example, for a dislocation loop the only important contribution for $q \gg 1/R_0$ comes from the region near the dislocation line. Treating the differential line segment as straight, we get in analogy to Ref. 21:

$$S_{\vec{h}}(\vec{q}) \sim c\,(N/V_c^2)\,[(bh)^2/q^5]R_0 . \tag{26}$$

For other cluster models, for which $\vec{t}(\vec{r})$ and $\delta\vec{h}(\vec{r})$ are bounded, we expect an even faster decrease. However, approximation (24), giving zero for such cases, is obviously no longer valid.

VI. MOMENTS OF DIFFUSE SCATTERING

By summing $S_{\vec{h}}(\vec{q})$ over all q values, we get the total integrated intensity of the diffuse scattering in the vicinity of the Bragg reflection $\vec{h}$. From Eq. (8) we obtain for this, using the Fourier presentation of the δ function and $\phi(\vec{R}=0) = 0$,

$$[V_c/(2\pi)^3] \int d\vec{q}\, S_{\vec{h}}(\vec{q}) = N\,(1 - e^{-2L_{\vec{h}}}) . \tag{27}$$

The second term $Ne^{-2L_{\vec{h}}}$ is the total integrated intensity of the Bragg scattering. Therefore, whereas for $L \ll 1$ the Bragg scattering dominates and the diffuse scattering is proportional to c, for larger L values the Bragg scattering disappears and the diffuse scattering contains the whole intensity. However, while the diffuse scattering can adequately be described by kinematical theory, Bragg scattering has to be calculated from dynamical theory. This yields for the total integrated reflection R:

$$R = R_{\text{Bragg}} + R_{\text{diffuse}} = R_p e^{-L_{\vec{h}}} + R_{1p}(1 - e^{-2L_{\vec{h}}}). \tag{28}$$

Here R_p is the integrated reflection for the "perfect" crystal, i.e., calculated by dynamical theory (neglecting absorption), and $R_{1p} \gg R_p$ is the value for the "imperfect" crystal (kinematical theory). Therefore R increases from R_p for $L \ll 1$ to R_{1p} for $L \gg 1$. The relation (28) can be used to determine the Debye-Waller factor.[15]

Similarly to the total diffuse scattering, we can also calculate certain moments of the diffuse scattering, generalizing a method of Wilson[23] for linewidth studies. For example, the first moment, the average of the jth component of $\vec{q}$, defined by

$$\langle q_j \rangle = \int d\vec{q}\, q_j S_{\vec{h}}(\vec{q}) / \int d\vec{q}\, S_{\vec{h}}(\vec{q}) , \tag{29}$$

can be calculated from (8) by replacing first q_j by a derivation with respect to R_j and then performing the $\vec{q}$ integration yielding the δ function $\delta(\vec{R})$. Thus we get

$$\langle q_j \rangle = \frac{1}{1 - e^{-2L_{\vec{h}}}} \left(e^{\phi(\vec{R})}\, i\, \frac{\partial \phi(\vec{R})}{\partial R_j} \right)_{\vec{R}=0} \equiv 0 . \tag{30}$$

In the same way we get expressions for higher moments, e.g., the second and third ones,

$$\langle q_i q_j \rangle = \frac{1}{1 - e^{-2L_{\vec{h}}}}\, c \int \frac{d\vec{r}}{V_c}\, \delta h_i(\vec{r})\, \delta h_j(\vec{r}) , \tag{31}$$

$$\langle q_i q_j q_k \rangle = \frac{1}{1 - e^{-2L_{\vec{h}}}}\, c \int \frac{d\vec{r}}{V_c}\, \delta h_i(\vec{r})\, \delta h_j(\vec{r})\, \delta h_k(\vec{r}) . \tag{32}$$

Again $\delta\vec{h}(\vec{r})$ [Eq. (24)] is the local change of the reciprocal-lattice vector $\vec{h}$. For small concentration $L \ll 1$, all moments are independent of the concentration, being consistent with the cluster form factor $G_{\vec{h}}(\vec{q})$ [Eq. (12)]. However, for larger concentrations ($e^{-2L} \ll 1$) the moments increase with the concentration. For example, $\langle \vec{q}^2 \rangle$ is proportional to c, resulting in a half-width proportional to $\sqrt{c}$. The third moment should reflect the asymmetry of the diffuse scattering. However, the sign of the third moment, being determined by the sign of $\delta\vec{h}(\vec{r})$, is opposite to the asymmetry observed in Eqs. (18) and (20) (see Sec. VII).

As an example, we may calculate these moments explicitly for a cluster of n_{cl} point defects distributed homogeneously within a sphere of radius R_0, the displacement field of which is

$$\vec{t}(\vec{r}) = \begin{cases} n_{cl} A\, \vec{r}/R_0^3 & \text{for } r < R_0 \\ n_{cl} A\, \vec{r}/r^3 & \text{for } r > R_0 . \end{cases} \tag{33}$$

For the moments in a certain direction $\vec{\kappa}$ ($\vec{\kappa}^2=1$) one gets

$$\langle(\vec{\kappa}\cdot\vec{q})^2\rangle=\frac{1}{1-e^{-2L}}c\frac{4\pi A^2 n_{cl}^2}{V_c R_0^3}\left[\frac{2}{15}(\vec{h}\cdot\vec{\kappa})^2+\frac{1}{15}h^2\right], \tag{34}$$

$$\langle(\vec{\kappa}\cdot\vec{q})^3\rangle=\frac{-1}{1-e^{-2L}}c\frac{4\pi A^3 n_{cl}^3}{V_c R_0^6}\left[\frac{6}{7}(\vec{h}\cdot\vec{\kappa})^3+\frac{24}{35}(\vec{h}\cdot\vec{\kappa})h^2\right]. \tag{35}$$

The moments of $S_{\vec{h}}(\vec{q})$ can give us also some information about the asymptotic behavior for large q values. For instance, for loops $S_{\vec{h}}(\vec{q})$ goes for large q as $1/q^5$ (26), and therefore the second moment diverges logarithmically. This can also be seen in Eq. (31) where the integral diverges logarithmically at the loop line. Similarly the second moment diverges for the asymptotic field $\vec{t}(\vec{r})$ $\sim 1/r^2$, but stronger than logarithmically, in agreement with the slower $1/q^4$ decreases of (25). On the other hand, the second and third moments for the field (33) exist but the fourth one diverges due to the discontinuity of $\delta\vec{h}(\vec{r})$ for $r=R_0$. Therefore $S_{\vec{h}}(\vec{q})$ will decrease as $1/q^6$. Thus the asymptotic behavior of $S_{\vec{h}}(\vec{q})$ is determined by the singularities, discontinuities, etc., of the displacement field in the core of the cluster.

VII. CENTER OF DIFFUSE SCATTERING

There is an apparent contradiction for the asymmetry following from the equations (18), (20), and (32), (35). Whereas according to (18), (20) the asymmetry for small q is opposite to the change of the Bragg reflection due to lattice expansion, the third moment (32) is directly proportional to the local change of $\vec{h}$. The first moment (30), on the other hand, vanishes. This discrepancy arises from the fact that the behavior of $S_{\vec{h}}(\vec{q})$ for large q enters into the moments with a much higher weight than the behavior for small or medium q's. Or in real space, the moments are essentially determined by the behavior of $\vec{t}(\vec{r})$ in the core of the cluster and not by the asymptotic field determining the Huang scattering. Therefore the moments do not necessarily give a fair picture of the major part of the diffuse scattering. To see this more clearly, we consider in the following the scattering from clusters with an "infinitesimal core," i.e., we assume that the displacement field is exactly given by the asymptotic formula $\vec{t}(\vec{r})\sim 1/r^2$.

Going back to (1) we identify, analogously to Krivoglaz, $\vec{R}^m$ as the positions in the lattice expanded homogeneously by the image forces alone. Further, we put $\vec{K}=\vec{h}^0+\vec{q}^0$ with $\vec{h}^0$ being the reciprocal-lattice vector of this lattice. $\vec{h}^0$ and $\vec{q}^0$ are connected with $\vec{h}$ and $\vec{q}$ (being related to the "average" lattice expanded by both real and image forces) by

$$\vec{h}=\vec{h}^0+\delta\vec{h} \quad \text{and} \quad \vec{q}=\vec{q}^0-\delta\vec{h}, \tag{36}$$

with

$$\delta\vec{h}=c\int\frac{d\vec{r}}{V_c}\,\delta\vec{h}(\vec{r})=-c\int\frac{d\vec{r}}{V_c}\,\partial_{\vec{r}}\,[\vec{h}\cdot\vec{t}(\vec{r})].$$

For the total intensity (diffuse and Bragg) we get from (1), replacing all sums by integrations as in Sec. II,

$$S_{\vec{h}^0}(\vec{q}^0)=N\int\frac{d\vec{R}}{V_c}e^{i\vec{q}^0\cdot\vec{R}}\langle e^{i\vec{h}^0\cdot[\vec{u}(\vec{R})-\vec{u}(0)]}\rangle$$

$$=N\int\frac{d\vec{R}}{V_c}e^{i\vec{q}\cdot\vec{R}}e^{\psi(\vec{R})}, \tag{37}$$

with

$$\psi(\vec{R})=c\int\frac{d\vec{R}}{V_c}\langle e^{i\vec{h}\cdot[\vec{t}(\vec{R}+\vec{r})-\vec{t}(\vec{r})]}-1\rangle.$$

Similarly to (30), the first moment of the total intensity is given by

$$\langle\vec{q}^0\rangle_{\text{total}}=-\langle\{\partial_{\vec{R}}\,[\vec{h}\cdot\vec{u}(\vec{R})]\}_{\vec{R}=0}\rangle=\{i\,\partial_{\vec{R}}\,\psi(\vec{R})\}_{\vec{R}=0}. \tag{38}$$

Therefore the average $\vec{q}$ value is essentially given by the ensemble average of the strain.

In order to get $\vec{q}^0$ we have to calculate $\psi(\vec{R})$ explicitly for small R. For the first derivative at $\vec{R}=0$ we need only the antisymmetric part $\psi^{(A)}$, which for small R is given by

$$\psi^{(A)}(\vec{R})\simeq ic\int\frac{d\vec{r}}{V_c}\,\sin\{\vec{R}\cdot\partial_{\vec{r}}\,[\vec{h}\cdot\vec{t}(\vec{r})]\}. \tag{39}$$

Now if $\vec{t}(\vec{r})$ and its derivative are finite everywhere, we can replace the sine function by the argument and get, with (36),

$$\psi^{(A)}(\vec{R})\simeq -i\,\delta\vec{h}\cdot\vec{R}$$

or

$$\langle\vec{q}^0\rangle_{\text{total}}=\delta\vec{h}=\langle\vec{q}^0\rangle_{\text{Bragg}}=\langle\vec{q}^0\rangle_{\text{diffuse}}, \tag{40}$$

which means that the total intensity and therefore also the diffuse intensity is centered at the Bragg reflection, in agreement with (30). However, for the infinitesimal cluster $\vec{t}(\vec{r})$ is singular at $\vec{r}=0$, leading, e.g., for

$$\vec{t}(\vec{r})=A\,\vec{r}/r^3,$$

to

$$\partial_{\vec{r}}\,[\vec{h}\cdot\vec{t}(\vec{r})]=\tfrac{4}{3}\pi A\,\delta(\vec{r})+A[\vec{h}/r^3-3(\vec{h}\cdot\vec{r})\vec{r}/r^5]. \tag{41}$$

For this case the replacement of the sine in (39) by the argument is not possible. But by introducing (41) into (39), the first term in (41) gives no contribution at all to the integral [because $|\sin\alpha\delta(\vec{r})| \leq 1$ and $=0$ for all $\vec{r} \neq 0$]. It is just this term which gives the only contribution to $\delta\vec{h}$ according to Eqs. (36) and (46). However, the integral with the second term of (41) vanishes for all $\vec{R}$ because its average over all directions is zero. This can be seen by substituting the whole second term as integration variable instead of $\vec{r}$. Then we get, by introducing the averages of the Bragg and diffuse scattering with the corresponding weight factors e^{-2L} and $1-e^{-2L}$,

$$\langle\vec{q}^0\rangle_{\text{total}} = 0 = \delta\vec{h}e^{-2L} + \langle\vec{q}^0\rangle_{\text{diffuse}}(1-e^{-2L})$$

or (42)

$$\langle\vec{q}^0\rangle_{\text{diffuse}} = -\delta\vec{h}e^{-2L}/(1-e^{-2L}) .$$

Whereas the center of the total intensity lies at the Bragg positions of the lattice expanded by the image forces alone, the center of the diffuse scattering is shifted from this position opposite to the change $\delta\vec{h}$ of the Bragg position due to the real forces. For small concentrations, $\langle\vec{q}^0\rangle_{\text{diffuse}}$ is much larger than $\delta\vec{h}$ and is equal to $\langle\vec{q}^0\rangle \simeq \langle\vec{q}\rangle \simeq -\delta\vec{h}/2L_{\vec{h}}$ independent of the concentration, which is in agreement with the asymmetry of (18) and (20). For $L \gg 1$ the Bragg intensities are negligible and $\langle\vec{q}^0\rangle_{\text{diffuse}} = 0$ whereas for medium L values $\langle\vec{q}^0\rangle_{\text{diffuse}}$ may be located by chance at the Bragg positions of the ideal unexpanded lattice, which seems to have been observed in Ref. 6.

Thus we have seen that for a cluster with an "infinitesimal core," the average $\langle\vec{q}^0\rangle_{\text{diffuse}}$ is drastically different from the average for a cluster with a finite core for which $\vec{t}(\vec{r})$ is finite everywhere. However, the averages for the latter ones are to a large extent determined by the scattering for very large q values, and hence are not representative for the major part of the scattering. Therefore, if we can define some modified moments, for which the scattering for large q is not so decisive, we expect for them essentially the same result as for the infinitesimal cluster. This can be done by introducing a function, e.g., $e^{-\epsilon q}$, which cuts off the intensity for large q values. However, ϵ has to be so small that practically all the intensity is contained in the region $q \leq 1/\epsilon$. So we have for the first moment of the diffuse intensity, starting as in (29) from the lattice expanded by the real and image forces,

$$\langle\vec{q}\rangle_\epsilon = \frac{\int d\vec{q}\,\vec{q}\,S_{\vec{h}}(\vec{q})e^{-\epsilon q}}{\int d\vec{q}\,S_{\vec{h}}(\vec{q})e^{-\epsilon q}}$$

$$= \frac{\int d\vec{R}\delta_\epsilon(\vec{R})e^{\phi(\vec{R})}\, i\,\partial_{\vec{R}}\,\phi(\vec{R})}{\int d\vec{R}\delta_\epsilon(\vec{R})\,(e^{\phi(\vec{R})} - e^{\phi(\infty)})} , \qquad (43)$$

with

$$\delta_\epsilon(\vec{R}) = \pi^{-2}\,\epsilon^2/(R^2+\epsilon^2)^2 \to \delta(\vec{R}) \quad \text{for } \epsilon \to +0 .$$

For $\epsilon \equiv 0$ we have the same result as (30). Choosing ϵ so that $\phi(\epsilon) \ll 1$, then the denominator is equal to $1-\epsilon^{-2L}$, stating that the total diffuse intensity is contained in the region $q \leq 1/\epsilon$. Then we get

$$\langle\vec{q}\rangle_\epsilon = \frac{-1}{1-e^{-2L}} \int d\vec{R}\,\delta_\epsilon(\vec{R})c$$

$$\times \int \frac{d\vec{r}}{V_c}\delta\vec{h}\cdot(\vec{R}+\vec{r})\{1-\cos\vec{h}\cdot[\vec{t}(\vec{R}+\vec{r})-\vec{t}(\vec{r})]\} , \qquad (44)$$

which shows that $\vec{q}$ is always opposite to the local change $\delta\vec{h}(\vec{r})$. Furthermore we assume that the displacements $\vec{t}_c$ in the core of the cluster are very large: $\vec{h}\cdot\vec{t}_c \gg 1$. Then the cosine function in (44) may oscillate even for relatively small values of $\vec{R}$, ϵ, respectively, and in the core of the cluster only the factor 1 remains. However, outside the core we get no contribution anyway because the angle integral of $\delta\vec{h}(\vec{r})$ vanishes there. More precisely, one can show, for instance in loops, that for $hb \gg 1$ and $L_{\vec{h}}/(hb)^{3/2} \approx c\,R_0^3/V_c \ll 1$, [17] one gets

$$\langle\vec{q}\rangle_\epsilon = -\delta\vec{h}/(1-\epsilon^{-2L_{\vec{h}}})$$

for all ϵ with $R_0 \gg \epsilon \gg R_0/hb$. (45)

Considering that $\vec{q}^0 = \delta\vec{h} + \vec{q}$, this is the same result as as for the infinitesimal cluster. This result is also clear, because if we make a cluster with a given asymptotic field compact enough ($R_0 \to 0$), then the conditions $\vec{h}\cdot\vec{t}_c \gg 1$ and $cR_0^3/V_c \ll 1$ are always fulfilled. However, the result (45) shows that the model of the infinitesimal cluster is reasonable only if the displacements in the core are very large, $\vec{h}\cdot\vec{t}_c \gg 1$. For small displacements, $\vec{h}\cdot\vec{t}_c \ll 1$, on the other hand, we see directly from (44) that $\langle\vec{q}\rangle_\epsilon \sim (\vec{h}\cdot\vec{t})^3$. Because $L \sim (\vec{h}\cdot\vec{t})^2$ in this case, we have practically no asymmetry.

For completeness we give here the values of $\delta\vec{h}$ for clusters with n_{cl} point defects assuming isotropy:

$$\delta\vec{h} = -c\,n_{cl}(4\pi A/3V_c)\vec{h} , \qquad (46)$$

and for dislocation loops [Burgers vector $\vec{b} = b\vec{b}^0$ perpendicular to the plane; $(\vec{b}^0)^2 = 1$, $b > 0$ for interstitial loops, $b < 0$ for vacancy loops]:

$$\delta\vec{h} = -c\frac{b\pi R_0^2}{V_c}\frac{2(\frac{4}{5}-\gamma)}{3(1-\gamma)}\vec{b}^0(\vec{b}^0\cdot\vec{h}) + \frac{\gamma-\frac{1}{5}}{3(1-\gamma)}\vec{h} . \qquad (47)$$

If we have only one kind of loop, then the reflections parallel to $\vec{b}^0$ are much more changed than the perpendicular ones.

For very high concentrations $L \gg 1$, Krivoglaz and Ryaboshapka[9,10] have given approximate solutions of the diffuse intensity. For loops, the resulting intensity forms for $(hb)^{3/2} \gg L \gg 1$ (essentially a Lorentz distribution) in agreement with (45), centered at the reciprocal-lattice positions of the lattice expanded by the image forces alone. For the opposite limit $L \gg (hb)^{3/2} \gg 1$, $c R_0^3/V_c \gg 1$ the distribution is Gaussian, centered at the positions of the Bragg reflections. However, for the latter case the concentration has to be so large that the loop planes are essentially close packed, which seems to be very unlikely.

VIII. CONCLUSIONS

The diffuse scattering from defect clusters, characterized by strong displacement fields, has been calculated. In Sec. I the general formulas for the diffuse scattering are given. The problems due to lattice expansion by the real and image forces are discussed, and it is shown that the procedure of Krivoglaz[9] is correct. For small concentrations the intensity is essentially determined by a "cluster form factor" describing the distorted lattice regions in the vicinity of the clusters. Due to the "coherent" action of all the defects in the same cluster, the intensity is extremely large near the Bragg reflection. For small $q \ll 1/R_0$ the Huang scattering $\sim 1/q^2$ dominates, giving information about the symmetry of the defects (Fig. 1). There is also an important asymmetry of the diffuse scattering, being always opposite to the change of the reflection vector due to lattice expansion (Fig. 2). It allows a simple determination whether interstitial-type or vacancy-type clusters are present. For large $q \gg 1/R_0$ the diffuse scattering decreases faster than $1/q^4$ and depends very much on the displacement field in the core of the cluster. In Secs. VI and VII simple expressions for the moments of the diffuse scattering are given. It is shown that for clusters with strong displacement fields the center of the diffuse scattering (i.e., the first moment) is systematically different from the position of the Bragg reflection. This explains the pronounced asymmetry of the diffuse scattering for relatively small concentrations, which has been observed recently,[12-16] as well as the shift of the maximum of the diffuse scattering for higher concentrations[10] which has been reported in Refs. 11 and 6.

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APPENDIX

In Ref. 15, rocking curves of the diffuse intensity have been obtained with the use of a double-crystal spectrometer. In this experiment one measures, with a wide-open counter, the total intensity scattered in the vicinity of a certain Bragg reflection as a function of the rocking angle $\delta\theta_B$ of the crystal, i.e., the angular deviation from the Bragg angle θ_B.

Therefore one has to integrate $S_{\vec{h}}(\vec{q})$ over all q values along the Ewald sphere. Since $q \approx 1/R_0$ is small, the Ewald sphere can be replaced by the tangential plane at the Bragg position. For a given rocking angle $\delta\theta_B$ we obtain a minimal q value of $q_0 = h \cos\theta_B \delta\theta_B$, and we have to integrate over all $\vec{q}'$ lying in the tangential plane $\vec{q}' \perp \vec{q}_0$, so that the measured intensity is given by

$$I_{\vec{h}}(\vec{q}_0) \sim \int_{\vec{q}' \perp \vec{q}_0} dF_{\vec{q}} S_{\vec{h}}(\vec{q}_0 + \vec{q}') \,. \tag{A1}$$

Similar to (15), $I_{\vec{h}}(\vec{q}_0)$ can be split up into a part $I_{\vec{h}}^S(\vec{q}_0)$ symmetrical with respect to $\vec{q}_0$ and an antisymmetrical part $I_{\vec{h}}^A(\vec{q}_0)$. Both parts can be calculated by replacing $S_{\vec{h}}$ by $S_{\vec{h}}^S$ or $S_{\vec{h}}^A$, respectively [Eq. (A1)].

From the behavior of $S_{\vec{h}}(\vec{q})$ for small q, (19) and (20), we obtain for $I_{\vec{h}}(\vec{q}_0)$ for $q_0 \ll 1/R_0$:

$$\begin{aligned} I_{\vec{h}}^S(\vec{q}_0) &\sim c\, N n_{cl}^2 \ln(q_{cl}/q_0); \\ I_{\vec{h}}^A(\vec{q}_0) &\sim c\, N n_{cl}^2 (q_0/q'_{cl}) \ln(q'_{cl}/q_0) \,. \end{aligned} \tag{A2}$$

Here $q_{cl} \approx 1/R_{cl} \approx q'_{cl}$ is the reciprocal of an average cluster radius R_{cl}. Therefore $I_{\vec{h}}^S(\vec{q}_0)$ diverges logarithmically for small q_0, whereas the asymmetry disappears.

Similarly, the behavior of $I_{\vec{h}}(\vec{q}_0)$ for large q_0 is determined by the asymptotic behavior of $S_{\vec{h}}(\vec{q})$ (Sec. V). For instance, if $S_{\vec{h}}(\vec{q})$ decreases $1/q^n$, with, e.g., $n = 4$ or 5, then $I_{\vec{h}}(\vec{q}_0)$ decreases $1/q^{n-2}$. Moreover, all moments of $S_{\vec{h}}(\vec{q})$ in the direction of $\vec{q}_0$ are identical with the moments of $I_{\vec{h}}(\vec{q}_0)$ for the same direction, since the $\vec{q}'$ integration in (A1) refers to the plane normal to $\vec{q}_0$.

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Ultrasonic Attenuation in Aluminum†

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The normal-state ultrasonic attenuation of longitudinal waves propagating along the principal symmetry directions in aluminum has been measured over a wide range of ql, where q is the phonon wave number and l is the electron mean free path. The usual quadratic frequency behavior at low ql and linear frequency behavior at high ql is observed for the electronic attenuation. At high ql the limiting values of attenuation divided by frequency are found to be strongly anisotropic and not in agreement with the free-electron prediction. Calculations assuming an isotropic deformation parameter with a pseudopotential representation of the Fermi surface show strong anisotropy, but the agreement with experiment is not good. From the pseudopotential Fermi surface generalized to include the effects of static strain, the anisotropy of the deformation tensor has been calculated. Using this model of the deformation, good agreement is achieved not only with the ultrasonic data but also with area changes observed from de Haas–van Alphen measurements under hydrostatic pressure.

I. INTRODUCTION

Ultrasonic waves propagating in a high-purity metal at low temperatures experience a large attenuation owing to the electrons in the metal. A general theory[1] expresses the electronic attenuation in terms of several integrals over the Fermi surface involving the wave number q, the electron mean free path l, and the deformation properties of the Fermi surface under static strain. At higher frequencies, where the product ql becomes much greater than 1, the attenuation becomes linear in the frequency and independent of electron mean free path. For longitudinal waves, the major contribution to the attenuation comes from those areas of the Fermi surface where the normal to the surface is nearly perpendicular to the direction of the propagation; these regions are known as effective zones. Since the longitudinal ultrasonic attenuation at high ql in a metal depends only upon the shape of its Fermi surface and upon the deformation properties along the effective zones, measurements in several different directions at high ql should provide information about the magnitude and anisotropy of the deformation tensor.

In addition to the availability of high-purity single crystals, aluminum is a desirable metal for such an investigation because its Fermi surface can be accurately expressed in terms of a pseudopotential model based on de Haas–van Alphen (dHvA) measurements.[2] Previous measurements[3–7] in aluminum have generally been over a limited range of ql and the high-frequency limiting values have not been in agreement. In this paper, measurements of longitudinal ultrasonic attenuation in aluminum over a wide range of ql are reported and the results