Effect of Defect Clustering on Anomalous X-Ray Transmission*

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A theory of anomalous x-ray transmission for crystals containing defect clusters of radius much less than an extinction distance is developed in a way analogous to the treatment of the thermal vibrations. A statistical distribution of the clusters is assumed, and only the coherent wave is considered. Two cluster models are discussed. The first represents a "loose" cluster of point defects, the second a dislocation loop. The absorption consists of two parts, the photoelectric absorption and the diffuse-scattering absorption. The photoelectric absorption is essentially given by a Debye-Waller factor resulting from the static lattice displacements of the clusters. The diffuse-scattering absorption is mainly determined by the very intense diffuse scattering in the wings of the Bragg reflection. Whereas for small clusters the photoelectric absorption is most important, for very large clusters the diffuse-scattering absorption dominates. Compared to the case of isolated point defects, the absorption can increase by as much as a factor of 10^5, if a given number of point defects cluster and, for instance, build dislocation loops.

I. INTRODUCTION

THE dynamical theory of x-ray diffraction by ideal crystals, in particular anomalous transmission, is well understood.[1] Dynamical theories for nonideal crystals have been developed recently; e.g., the thermal motion of the lattice atoms has been treated by several authors.[2–4] These theories consider only the mean elastic or coherent wave, the scattering of which can be described by an effective periodic potential. The same theory can also be generalized and applied to crystals with statistically distributed point defects.[5,6] In this paper, we will develop the theory one step further and apply it to statistically distributed defect clusters, the sizes of which are small compared with an extinction length. We will discuss the effects on anomalous transmission which result if point defects cluster together.

Such effects were first observed by Patel and Batterman,[7] who found a two order of magnitude decrease of the Borrmann intensity resulting from the clustering of oxygen in dislocation-free Si. Efimov and co-workers[8,9] have published a series of papers about their investigation of the effect of impurities, vacancies, and clusters on the Borrmann intensity in Ge crystals. They found that even an extremely small concentration of hydrogen impurities ($\sim 10^{14}$ cm^{-3}) and of precipitated copper and nickel impurities ($\sim 10^{16}$ cm^{-3}) can cause a considerable decrease of the Borrmann intensities. The most detailed experiment has been done recently by Baldwin *et al.*,[10] who investigated the anomalous transmission through neutron-irradiated Cu crystals. They found large changes in the Borrmann intensity after irradiation at 40°C; the effective absorption was a factor of 10 to 20 larger than one would expect for a reasonable concentration of point defects. Moreover, they observed a different wavelength dependence of the absorption than the photoelectric one. Their experiment suggested strongly that the exact form of the defects, i.e., whether randomly distributed point defects or defect clusters, must be very important.

II. THEORY OF COHERENT WAVE

Our theory applies only to a statistical defect distribution; that means we assume that the measured intensity does not depend on the accidental microscopic defect configuration in a given crystal, but only on macroscopic quantities as the average defect density, their pair correlations, etc. Therefore, we average the measured intensity over all possible microscopic defect configurations. Significant deviations from this average are not expected for crystals with very many defects.

As for the temperature theory,[2–4] we assume that for the Bragg-reflected intensities we can restrict ourselves to the coherent wave, being that part of the total wave which interferes with the incident wave. This coherent wave is given by the average of the total wave $\mathbf{D}(\mathbf{r})$ over all microscopic defect configurations leading to the same macroscopic situation: $\mathbf{D}_{\mathrm{coh}}(\mathbf{r})=\langle\mathbf{D}(\mathbf{r})\rangle$.[6] The other parts of the total wave, the diffuse waves, do not satisfy the Bragg condition because they are scattered by an oblique angle due to the transfer of a pseudomomentum from the defect lattice (phonon momentum in the tem-

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[8] O. N. Efimov and A. M.Elistratov, Fiz. Tverd. Tela **5**, 1869 (1963) [English transl.: Soviet Phys.—Solid State **5**, 1364 (1963)]; *ibid.* **5**, 2116 (1963) [English transl.: *ibid.* **5**, 1543 (1963)].
[9] O. N. Efimov and L. I. Datsenko, Fiz. Tverd. Tela **8**, 809 (1966) [English transl.: Soviet Phys.—Solid State **8**, 648 (1966)].
[10] T. O. Baldwin, F. A. Sherrill, and F. W. Young, J. Appl. Phys. **39**, 1541 (1968).

perature case). Therefore, in the Borrmann case, they are absorbed in the normal way and have no chance to penetrate thick crystals. The hypothesis that all the Bragg intensities are given by the coherent wave clearly breaks down for defect clusters large with respect to an extinction length, for then the diffuse waves are scattered by a very small angle and partially satisfy the Bragg condition, leading to line broadening in the Bragg case. Therefore, we restrict ourselves to point defects and small clusters, whose radii are small compared with the extinction length.

The incident electrical displacement field $\mathbf{D}_0(\mathbf{r})=\mathbf{D}_0e^{i\mathbf{k}_0\cdot\mathbf{r}}$ is scattered by a crystal with a specific configuration of self-defects (vacancies and interstitials). For simplicity, we will describe the interaction by a susceptibility $\chi(\mathbf{r})$, being complex to include the photoelectric absorption. Moreover, we choose a rigid ion model.

$$\chi(\mathbf{r})=-(4\pi/k_0^2)(e^2/m)\rho(\mathbf{r})=\chi'(\mathbf{r})+i\chi''(\mathbf{r})$$
$$=\sum_n \xi(\mathbf{r}-\mathbf{R}^n),\quad \xi=\xi'+i\xi''. \tag{1}$$

Here, $\rho(\mathbf{r})$ is the (complex) electron density and $\xi(\mathbf{r}-\mathbf{R}^n)$ is the atomic susceptibility of an atom at the position $\mathbf{R}^n$. In principle, such a classical description of the photoelectric absorption does not apply, because the electrons have to be treated quantum mechanically.[11–13] As Molière[12] has shown, one gets instead of (1) a generalized nonlocal susceptibility, determined by the induced-current correlations in the lattice atoms. However, due to the localization of the atomic orbitals, this term can also be split into single-atom contributions so that the following treatment is not changed essentially. Due to the defects, $\mathbf{R}^n=\bar{\mathbf{R}}^n+\mathbf{s}^n$ consists of a position $\bar{\mathbf{R}}^n$ in the (expanded) "average" lattice and a displacement $\mathbf{s}^n$ from this lattice site. Therefore, $\chi(\mathbf{r})$ is no longer periodic. The total field $\mathbf{D}(\mathbf{r})$ is determined by the Maxwell's equations which are, in von Laue's form,

$$(\nabla_r^2+k_0^2)\mathbf{D}(\mathbf{r})=-\nabla_r\times\nabla_r\times[\chi(\mathbf{r})\mathbf{D}(\mathbf{r})]. \tag{2}$$

$\mathbf{D}(\mathbf{r})$ depends parametrically on all the defect configurations. By averaging $\mathbf{D}(\mathbf{r})$, we obtain the coherent field $\langle\mathbf{D}(\mathbf{r})\rangle$. It can be shown[3] that $\langle\mathbf{D}\rangle$ obeys an equation like (2) with an effective operator $\mathbf{U}$, having the great advantage of being periodic.

$$(\nabla_r^2+k_0^2)\langle\mathbf{D}(\mathbf{r})\rangle=\int d\mathbf{r}'\,\mathbf{U}(\mathbf{r},\mathbf{r}')\cdot\langle\mathbf{D}(\mathbf{r}')\rangle. \tag{3}$$

The operator $\mathbf{U}$, being a tensor, is in general nonlocal, non-Hermitian, and energy-dependent, and reflects all the defect properties of the lattice. $\mathbf{U}=\mathbf{R}+i\mathbf{I}$ can be split up into an Hermitian part $\mathbf{R}$, being responsible for the scattering, and an anti-Hermitian part $\mathbf{I}$, describing the absorption.

In a first approximation, $\mathbf{U}=\mathbf{U}^{(1)}+\mathbf{U}^{(2)}\simeq\mathbf{U}^{(1)}$ is local and essentially given by the averaged susceptibility, the imaginary part ($\mathbf{I}^{(1)}$) of which gives *the photoelectric absorption*

$$\mathbf{U}^{(1)}(\mathbf{r},\mathbf{r}')=-\nabla_r\times\nabla_r\times\langle\chi(\mathbf{r})\rangle\delta(\mathbf{r}-\mathbf{r}')$$
$$=[\mathbf{R}^{(1)}(\mathbf{r})+i\mathbf{I}^{(1)}(\mathbf{r})]\delta(\mathbf{r}-\mathbf{r}'). \tag{4}$$

In this approximation, we have totally neglected the influence of the diffuse scattering. This can be taken into account by a correction $\mathbf{U}^{(2)}$, the imaginary term $\mathbf{I}^{(2)}$ of which describes the *absorption* of the coherent wave due to the production of *diffuse waves*. As we will see later, this absorption is very important in the case of defect clustering, whereas it is small in the case of isolated point defects[6] or in the temperature case.[3,4]

$$\mathbf{U}^{(2)}(\mathbf{r},\mathbf{r}')$$
$$=\nabla_r\times\nabla_r\times\langle\delta\chi'(\mathbf{r})G(\mathbf{r},\mathbf{r}')\nabla_{r'}\times\nabla_{r'}\times\delta\chi'(\mathbf{r}')\rangle, \tag{5}$$

with

$$G(\mathbf{r},\mathbf{r}')\approx-(1/4\pi)e^{ik_0|\mathbf{r}-\mathbf{r}'|}/|\mathbf{r}-\mathbf{r}'|$$

and $\delta\chi'(\mathbf{r})=\chi'(\mathbf{r})-\langle\chi'(\mathbf{r})\rangle$. Here the replacement of the dynamic Green's function $G(\mathbf{r},\mathbf{r}')$ by the free Green's function is allowed only if the long-range correlations $\langle\delta\chi'(\mathbf{r})\delta\chi'(\mathbf{r}')\rangle$ are cut off for distances $|\mathbf{r}-\mathbf{r}'|$ larger than the extinction length.[3,4] Due to the periodicity of $\mathbf{U}$ $[\mathbf{U}(\mathbf{r},\mathbf{r}')=\mathbf{U}(\mathbf{r}+\bar{\mathbf{R}}^n,\mathbf{r}'+\bar{\mathbf{R}}^n)]$, the eigenfunctions of (3) are Bloch waves

$$\langle\mathbf{D}(\mathbf{r})\rangle=\frac{1}{\sqrt{V}}\sum_{\mathbf{h},s=1,2}e^{i(\mathbf{k}+\mathbf{h})\cdot\mathbf{r}}\mathbf{e}_\mathbf{h}^s D_\mathbf{h}^s, \tag{6}$$

with $\mathbf{e}_\mathbf{h}^s\cdot\mathbf{e}_\mathbf{h}^{s'}=\delta_{ss'}$ and $\mathbf{e}_\mathbf{h}^s\cdot(\mathbf{k}+\mathbf{h})=0$; $s,s'=1,2$. Here, $\mathbf{h}$ are the reciprocal-lattice vectors, and $\mathbf{e}_\mathbf{h}^s$ are the two polarization-unit vectors perpendicular to $\mathbf{k}+\mathbf{h}$. With (6), we get from (3) the fundamental equations determining the fields in the crystal,

$$[k_0^2-(\mathbf{k}+\mathbf{h})^2]D_\mathbf{h}^s=\sum_{\mathbf{h}',s'}U_{\mathbf{h}\mathbf{h}'}^{ss'}D_{\mathbf{h}'}^{s'}, \tag{7}$$

with

$$U_{\mathbf{h}\mathbf{h}'}^{ss'}=\frac{1}{V}\int_V d\mathbf{r}d\mathbf{r}'e^{-i(\mathbf{k}+\mathbf{h})\cdot\mathbf{r}}\mathbf{e}_\mathbf{h}^s\cdot\mathbf{U}(\mathbf{r},\mathbf{r}')\cdot\mathbf{e}_{\mathbf{h}'}^{s'}e^{i(\mathbf{k}+\mathbf{h}')\cdot\mathbf{r}'}$$
$$=R_{\mathbf{h}\mathbf{h}'}^{ss'}+iI_{\mathbf{h}\mathbf{h}'}^{ss'}. \tag{8}$$

To calculate the eigenfunctions of (7) for the two-beam case ($\mathbf{k}$ and $\mathbf{k}+\mathbf{h}$) we take, in a first approximation, only the real part $\mathbf{R}^{(1)}$ of $\mathbf{U}^{(1)}$ (4) into account. If the usual choice is made for the polarization vectors [for $s=1$ (σ or $\perp$ polarization), $\mathbf{e}_0^1=\mathbf{e}_\mathbf{h}^1$ perpendicular to both $\mathbf{k}$ and $\mathbf{k}+\mathbf{h}$; for $s=2$ (π or $\parallel$ polarization), $\mathbf{e}_0^2$ and $\mathbf{e}_\mathbf{h}^2$ in the ($\mathbf{k}$, $\mathbf{k}+\mathbf{h}$) plane with $\mathbf{e}_0^2\cdot\mathbf{e}_\mathbf{h}^2=\cos2\theta_B$], then the equations for the σ and π polarizations decouple. We get four dispersion surfaces, two for each polarization. The absorption due to $\mathbf{I}$ can then be calculated by per-

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[12] G. Molière, Ann. Physik **35**, 272 (1939); **35**, 313 (1939).
[13] H. Wagenfeld, Phys. Rev. **144**, 216 (1966).

turbation theory and is essentially given by the expectation value of $\mathbf{I}$ in the eigenstates.[3] For the symmetrical Laue case we get

$$\mu=-(1/k)[\tfrac{1}{2}(I_{00}{}^{ss}+I_{\mathbf{hh}}{}^{ss}) \pm W(1+W^2)^{-1/2}\,\tfrac{1}{2}(I_{00}{}^{ss}-I_{\mathbf{hh}}{}^{ss}) \mp(1+W^2)^{-1/2}\,\tfrac{1}{2}(I_{0\mathbf{h}}{}^{ss}+I_{\mathbf{h}0}{}^{ss})] \quad (9)$$

with

$$W=-(\mathbf{k}+\tfrac{1}{2}\mathbf{h})\cdot\mathbf{h}/Pk^2\chi_{\mathbf{h}}e^{-M_{\mathbf{h}}}$$

and

$$p=1, \quad \text{for } \sigma(s=1)\text{ pol}$$
$$=|\cos 2\theta_B|, \quad \text{for } \pi(s=2)\text{ pol.} \quad (10)$$

The upper sign in (9) is valid for the weakly absorbed wave fields and the lower one for the strongly absorbed ones. Now we assume that our lattice defects are self-interstitials on the interstitial position $\mathbf{R}^I$ in the unit cell. Then the susceptibility can be written as

$$\chi(\mathbf{r})=\sum_n \xi(\mathbf{r}-\bar{\mathbf{R}}^n-\mathbf{s}^n)+\sum_n p_n{}^I\xi(\mathbf{r}-\bar{\mathbf{R}}^n-\mathbf{R}^I-\mathbf{s}_I{}^n), \quad (11)$$

with a random number $p_n{}^I$, where

$$p_n{}^I=(p_n{}^I)^2=1 \quad \text{if interstitial position } (\bar{\mathbf{R}}^n+\mathbf{R}_I) \text{ is occupied}$$
$$=0 \quad \text{if interstitial position } \bar{\mathbf{R}}^n+\mathbf{R}_I \text{ is vacant.} \quad (12)$$

The average $\langle p_n{}^I\rangle_{\text{av}}$ is just the interstitial concentration c_I. With (11), we get for the Fourier coefficients $U_{\mathbf{hh}'}{}^{(1)ss'}$ (8) from (4)

$$U_{\mathbf{hh}}{}^{(1)ss'}=-\mathbf{e}_{\mathbf{h}}{}^s\cdot\mathbf{e}_{\mathbf{h}'}{}^{s'}(\mathbf{k}+\mathbf{h})^2\chi_{\mathbf{h}-\mathbf{h}'} \times(e^{-L_{\mathbf{h}-\mathbf{h}'}}+c_Ie^{-i(\mathbf{h}-\mathbf{h}')\cdot\mathbf{R}^I}e^{-L_{\mathbf{h}-\mathbf{h}'^I}}), \quad (13)$$

with

$$(\mathbf{k}+\mathbf{h})^2\chi_{\mathbf{h}-\mathbf{h}'}=\frac{(\mathbf{k}+\mathbf{h})^2}{V_c}\int_{-\infty}^{\infty}d\mathbf{r}e^{-i(\mathbf{h}-\mathbf{h}')\cdot\mathbf{r}}\xi(\mathbf{r})$$
$$=+\gamma f_{\mathbf{h}-\mathbf{h}'}, \quad \gamma=-(4\pi/V_c)e^2/mc^2.$$

Here $f_{\mathbf{h}-\mathbf{h}'}=f_{\mathbf{h}-\mathbf{h}'}{}'+if_{\mathbf{h}-\mathbf{h}'}{}''$ is the complex atomic scattering factor and V_c is the volume of the unit cell. $L_{\mathbf{h}}$ is a Debye-Waller factor due to the interstitial displacement field, defined by

$$e^{-L_{\mathbf{h}}}=\langle e^{-i\mathbf{h}\cdot\mathbf{R}^n}\rangle=\langle e^{-i\mathbf{h}\cdot\mathbf{s}^n}\rangle, \quad \mathbf{R}^n=\bar{\mathbf{R}}^n+\mathbf{s}^n. \quad (14)$$

Introducing the imaginary part $I^{(1)}$ of (4) into (9), we get for the photoelectric absorption

$$\mu^{PE}=\frac{\gamma}{k}\left(f_0{}''\mp\frac{1}{(1+W^2)^{1/2}}Pf_{\mathbf{h}}{}''e^{-L_{\mathbf{h}}}\right) +c_I\frac{\gamma}{k}\left(f_0{}''\mp\frac{\cos\mathbf{h}\cdot\mathbf{R}^I}{(1+W^2)^{1/2}}Pf_{\mathbf{h}}{}''e^{-L_{\mathbf{h}}{}^I}\right). \quad (15)$$

As a function of W, μ^{PE} shows the well-known dip in the absorption for the weakly absorbed wave fields (upper curve in Fig. 1). Assuming that $f_0{}''\approx f_{\mathbf{h}}{}''$ and $L_{\mathbf{h}}\ll 1$, we

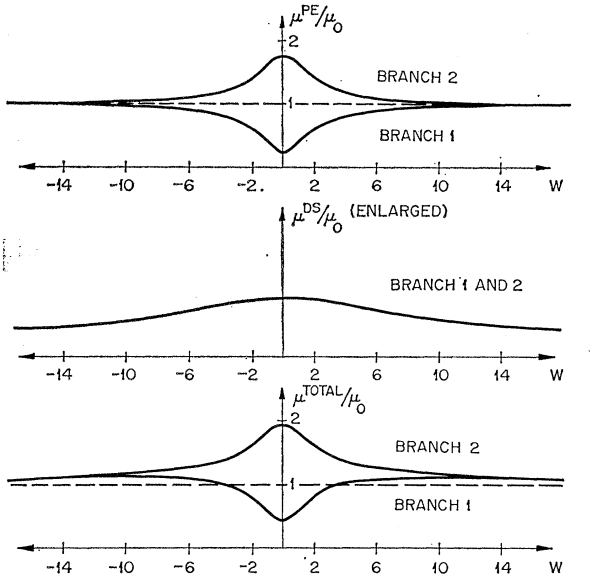

FIG. 1. Photoelectric absorption μ^{PE}, diffuse scattering absorption μ^{DS}, and total absorption μ^{total} as a function of the parameter W.

get for the σ-polarized and weakly absorbed wave field ($W=0$)

$$\mu^{\text{PE}}\approx\mu_0L_{\mathbf{h}}+c_I\mu_0(1-\cos\mathbf{h}\cdot\mathbf{R}^I),$$

with

$$\mu_0=(\gamma/k)f_0{}''. \quad (16)$$

The absorption consists of two parts. The first one, given essentially by $L_{\mathbf{h}}$, is the photoelectric absorption of the lattice atoms which are displaced by the interstitials into the space between the reflecting lattice planes and, therefore, absorb the Borrmann wave more strongly. The second term is the photoelectric absorption of the interstitials themselves, being largest if the interstitials are located in the middle between the reflecting planes ($\cos\mathbf{h}\cdot\mathbf{R}^I=-1$). For randomly distributed interstitials, both terms are of the same order of magnitude, e.g., in Cu.[6,14] If the interstitials are clustered together, the last term does not change as long as the interstitial position $\mathbf{R}^I$ remains the same. But we will see in the following that the Debye-Waller factor can increase remarkably due to clustering and so can the photoelectric absorption of the lattice atoms.

In the case of vacancies, the second part in (15) and (16) is missing and the first part is multiplied by a factor $1-c_v\simeq 1$. The "imaginary" part $\mathbf{I}^{(2)}$ of $\mathbf{U}^{(2)}(5)$ describes the "diffuse scattering absorption." Analogously to the photoelectric absorption, the diffuse scattering consists also of two parts, one part due to the scattering at the displacement correlations of the lattice atoms, and a second part due to a direct scattering at the defects (e.g., interstitials). In the following, it will be shown that the scattering at the displaced lattice atoms is very sensitive

[14] G. C. North and G. J. Ring (private communication).

to defect clustering and can increase several orders of magnitude, depending on the cluster radius. The direct scattering at the defect atoms is much less sensitive to clustering and will, therefore, be neglected here. Then the Fourier coefficients $I^{(2)}{}_{\mathbf{h}\mathbf{h}'}{}^{ss'}$ can be expressed as a surface integral $dF_{\mathbf{k}'}$ over the Ewald sphere $\mathbf{k}'^2=k_0^2$:

$$I^{(2)}{}_{\mathbf{h}\mathbf{h}'}{}^{ss'}=-\left(\frac{e^2}{mc^2}\right)^2\frac{1}{V_c k_0}\int_{k'^2=k_0^2} dF_{\mathbf{k}'} f_{\mathbf{k}+\mathbf{h}-\mathbf{k}'} f_{\mathbf{k}'-\mathbf{k}-\mathbf{h}'} \times(\mathbf{e}_{\mathbf{h}}{}^{s}\cdot\mathbf{e}_{\mathbf{h}'}{}^{s'}-(\mathbf{e}_{\mathbf{h}}{}^{s}\cdot\mathbf{k}'/k')(\mathbf{e}_{\mathbf{h}'}{}^{s'}\cdot\mathbf{k}'/k')) \times S_{\mathbf{h}\mathbf{h}'}(\mathbf{k}-\mathbf{k}'), \quad (17)$$

with

$$S_{\mathbf{h}\mathbf{h}'}(\mathbf{k}-\mathbf{k}') =\sum_m \exp i\mathbf{q}\cdot(\bar{\mathbf{R}}^m-\bar{\mathbf{R}}^{m'})\left(\langle e^{-i(\mathbf{k}+\mathbf{h}-\mathbf{k}')\cdot\mathbf{s}^m+i(\mathbf{k}+\mathbf{h}'-\mathbf{k}')\cdot\mathbf{s}^{m'}}\rangle -\langle e^{-i(\mathbf{k}+\mathbf{h}-\mathbf{k}')\cdot\mathbf{s}^m}\rangle\langle e^{i(\mathbf{k}+\mathbf{h}'-\mathbf{k}')\cdot\mathbf{s}^{m'}}\rangle\right) \quad (18)$$

with

$$\mathbf{k}'=\mathbf{k}+\mathbf{g}+\mathbf{q}.$$

Here, $\mathbf{q}$ is a vector within the *first Brillouin zone* and $\mathbf{g}$ is a reciprocal-lattice vector; both are chosen so that $\mathbf{k}'-\mathbf{k}=\mathbf{g}+\mathbf{q}$. Due to the translational invariance, the first average in (18) depends only on $\bar{\mathbf{R}}^m-\bar{\mathbf{R}}^{m'}$. All the information about the distorted lattice is contained in $S_{\mathbf{h}\mathbf{h}'}$.

III. TWO MODELS FOR DEFECT CLUSTERING

In order to discuss the effect of defect clustering on absorption, we have to know something about the displacement fields of such defect aggregates. Because calculations from first principles are not available, we make two models for defect clustering.

A. Cluster Model

We assume that the point defects are clustered together such that their displacement fields simply superimpose on each other. This would be the case for a very loose cluster with a small defect concentration. Furthermore, we will assume that the cluster centers are statistically distributed and that the clusters have spherical symmetry.

For the displacement fields $\mathbf{t}(\mathbf{r})$ of the single point defects, we apply continuum theory which gives us for the case of three equal crossed dipole forces P_0:

$$t_i(\mathbf{r})=-P_0\sum_j \nabla_{r_j}G_{ij}(\mathbf{r}),$$

with

$$G_{ij}(\mathbf{r})=(1/4\pi r)g_{ij}(\mathbf{r}/r). \quad (19)$$

Here, the elastic Green's function $G_{ij}(\mathbf{r})$ gives the displacements in i direction due to a unit force at the origin in j direction.[15,16] In an isotropic medium ($c_{11}=c_{12}+2c_{44}$), we get from (19)

$$t_i(\mathbf{r})=Ar_i/r^3, \quad \text{with} \quad A=P_0/[4\pi(c_{12}+2c_{44})]. \quad (20)$$

To discuss the influence of anisotropy, we use Leibfried's perturbation theory for weak anisotropy.[16,17] In the cubic coordinate system, we get

$$t_i(\mathbf{r})=\frac{P_0}{4\pi(c_{12}{}^v+2c_{44}{}^v)}\frac{1}{r^2}\left\{\rho_i+\frac{d}{c_{44}{}^v}\left[\tfrac{3}{2}(1-\beta)\rho_i{}^3 +\frac{15}{8}\beta\rho_i\sum_m\rho_m{}^4-\left(\frac{9}{10}+\frac{9}{40}\right)\beta\rho_i\right]\right\}, \quad (21)$$

with $\rho_i=r_i/r$, $c_{12}{}^v=c_{12}+d/5$, $c_{44}{}^v=c_{44}+d/5$,

$$\beta=(c_{12}{}^v+c_{44}{}^v)/(c_{12}{}^v+2c_{44}{}^v),$$

and $d=c_{11}-c_{12}-2c_{44}$ ($d=0$ for isotropy).

The volume change due to a distribution of dipole moments is in general given by (S_{ijkl}=inverse of the elasticity tensor C_{ijkl})[15]

$$\Delta V=\sum_{ikl} S_{iikl}P_{kl}, \quad (22)$$

where P_{kl} is the total dipole moment. Therefore, the volume change of a point defect with $P_{kl}=P_0\delta_{kl}$ is

$$\Delta V=3P_0/(3c_{12}+2c_{44}) =4\pi A3(1-\nu)/(1+\nu) \text{ for the isotropic case.} \quad (23)$$

Due to the superposition approximation, the volume change of a cluster of n_{cl} defects is just $n_{\mathrm{cl}}\cdot\Delta V$. Therefore, the volume of the crystal and the lattice constants of the average lattice are the same whether the defects are isolated or clustered.

B. Loop Model

For higher defect densities within the cluster, the nonlinear interaction and relaxation of the defects is very important, e.g., the point defects may collapse into a flat disk and form a dislocation loop. In this case, continuum theory can give us again the displacement field, which is produced by a distribution of dipole moments $p_{jk}=\sum_{l,m}C_{jklm}b_l dF_m$ on the loop plane F (b=Burgers vector, $\mathbf{F}$=vector normal to the loop plane $F=|\mathbf{F}|$). The displacement field is[17]

$$t_i(\mathbf{r})=-\sum_{jklm}\int_F \nabla_{r_k}G_{ij}(\mathbf{r}-\mathbf{r}')C_{jklm}b_l dF_m{}'\equiv\tau_i\left(\frac{\mathbf{r}}{R_0}\right). \quad (24)$$

Due to the radial dependence of $\nabla_{r_k}G_{ij}(\mathbf{r})\sim 1/r^2$, the displacement field of a loop with average radius R_0 depends only on the reduced coordinate $\mathbf{r}/R_0$. For an isotropic crystal the displacement field of a circular loop has been given by Kroupa[18] and Bullough *et al.*[19] in

[15] For a recent review, see R. Siems, KFA-JUEL Report No. JUEL-545-FN, 1968 (unpublished).
[16] P. H. Dederichs and G. Leibfried (unpublished).
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[18] F. Kroupa, Czech. J. Phys. **B10**, 284 (1960).
[19] R. Bullough and R. C. Newman, Phil. Mag. **5**, 921 (1960).

terms of elliptic integrals, and series expansions have been discussed by Keating and Goland.[20] In the following, we are mainly interested in the asymptotic displacement field for large r, which can be obtained in the general case from (24):

$$t_i(\mathbf{r}) \simeq -\sum_{j,k} \nabla_{r_k} G_{ij}(\mathbf{r}) P_{jk},$$

with

$$P_{jk} = \sum_{l,m} C_{jklm} b_l F_m. \tag{25}$$

In the isotropic case, we have for a not necessarily circular loop ($F=\pi R_0^2$), but with $\mathbf{b}$ parallel to $\mathbf{F}$,[18,21]

$$\mathbf{t}(\mathbf{r}) = \frac{bR_0^2}{8r^2}\left(\frac{1-2\nu}{1-\nu}[2\mathbf{b}^0(\mathbf{b}^0\cdot\boldsymbol{\varrho})-\boldsymbol{\varrho}] + \frac{3}{1-\nu}\boldsymbol{\varrho}(\mathbf{b}^0\cdot\boldsymbol{\varrho})^2\right), \tag{26}$$

with $\boldsymbol{\varrho}=\mathbf{r}/r$, $\mathbf{b}^0=\mathbf{b}/|\mathbf{b}|$, and $b=\pm|\mathbf{b}|$ for interstitial or vacancy loops. The volume change can be calculated from (22) with P_{jk} of Eq. (25).

$$\Delta V = \sum_{i,k,l,m,n} S_{iikl} C_{klmn} b_m F_n = \mathbf{b}\cdot\mathbf{F}. \tag{27}$$

The ΔV is independent of the loop shape and the anisotropy and equal to the volume occupied by the defects which build up the loop. It is opposite for interstitial and vacancy loops. Therefore, equal numbers of interstitials and vacancy loops give no net volume expansion,[22] in contrast to the fact that isolated interstitials and vacancies give a considerable expansion. Also, in this model we assume that the centers of the dislocation loops are randomly distributed.

The common feature of both cluster models is that the asymptotic displacement field of an aggregate of n_{cl} defects is proportional to n_{cl}/r^2, which will be most important for the anomalous absorption. Therefore, other cluster models which are associated with such a strong displacement field will give qualitatively the same result (e.g., small impurity precipitates), whereas clusters without strong displacement fields (as may be the case for voids) will show no large effects on anomalous transmission. The main difference between the two models is that for the loose clusters we neglect any relaxation effect of the interacting point defects, whereas this relaxation is taken into account in the loop model representing a dense cluster. Further, the clusters are assumed to have spherical symmetry, whereas the loops are highly anisotropic in shape.

IV. DEBYE-WALLER FACTOR FOR CLUSTER MODEL

Assuming that the static displacement $\mathbf{s}$ of a lattice atom due to the various point defects has a Gaussian distribution, we get from (9) for the Debye-Waller factor (of cubic crystals)

$$L_{\mathrm{h}} = \tfrac{1}{2}\langle(\mathbf{h}\cdot\mathbf{s})^2\rangle = \tfrac{1}{6}h^2\langle\mathbf{s}^2\rangle. \tag{28}$$

In the cluster model, the displacement $\mathbf{s}$ is the sum of displacement fields $\mathbf{t}_i$ due to all the point defects $i=1, 2, \ldots, N_d$. If the point defects are statistically independently distributed, we get

$$\langle\mathbf{s}^2\rangle = \sum_{i,j=1}^{N_d}\langle\mathbf{t}_i\cdot\mathbf{t}_j\rangle = \sum_{i=1}^{N_d}\langle\mathbf{t}_i^2\rangle = N_d\langle\mathbf{t}_i^2\rangle, \tag{29}$$

because $\langle\mathbf{t}_i\cdot\mathbf{t}_j\rangle=0$ for $i\neq j$ and because each defect i gives the same contribution by averaging over its position. The result is linear in N_d, which can be expressed by saying that the displacements $\mathbf{t}_i$ of different point defects add incoherently (addition of $\mathbf{t}_i^2$).

If the point defects are clustered together and form N_{cl} clusters, each containing n_{cl} defects ($N_d=N_{\mathrm{cl}}\cdot n_{\mathrm{cl}}$), and moreover if the cluster radii are small, then

$$\langle\mathbf{s}^2\rangle = \sum_{i,j=1}^{N_d}\langle\mathbf{t}_i\cdot\mathbf{t}_j\rangle = n_{\mathrm{cl}}\sum_{i=1}^{N_d}\langle\mathbf{t}_i^2\rangle$$
$$= N_d\cdot n_{\mathrm{cl}}\langle\mathbf{t}_i^2\rangle = N_{\mathrm{cl}}n_{\mathrm{cl}}^2\langle\mathbf{t}_i^2\rangle, \tag{30}$$

because each cluster, which contains the defect i, contains also $n_{\mathrm{cl}}-1$ other defects j with the same displacement field $\mathbf{t}_j=\mathbf{t}_i$. Therefore, the displacements of all the defects in the same cluster add coherently, whereas the different clusters add incoherently. Comparing (30) with (29), we see that the Debye-Waller factor L_{h} and also the absorption μ^{PE} increases by a factor n_{cl} due to the clustering.

Actually, Eq. (30) overestimates the clustering effect, because we assumed the cluster radius $R_{\mathrm{cl}}\approx 0$. For clusters with finite radii, the displacements of the point defects in the same cluster no longer add strictly coherently. To see this, we write by using the random numbers p_n of (12), $\mathbf{s}^m=\sum_n p_n\mathbf{t}^{m-n}$ and get

$$\langle(\mathbf{s}^m)^2\rangle = \sum_{n,n'}\langle p_n p_{n'}\rangle\mathbf{t}^{m-n}\cdot\mathbf{t}^{m-n'}. \tag{31}$$

Here $\langle p_n p_{n'}\rangle$ is the probability of finding one defect at position n and another one at position n'. It can be written as

$$\langle p_n p_{n'}\rangle = \delta_{nn'}c + (1-\delta_{nn'})[c^2 + cg(n-n')]. \tag{32}$$

The $g(n-n')$ is the conditional probability of finding one defect at position n, if there is already a defect of the *same* cluster at position n'. The g is normalized to $\sum_{n\neq 0} g(n) = n_{\mathrm{cl}}-1\approx n_{\mathrm{cl}}$ and vanishes, if the point defects are randomly distributed. With (32), we get

[20] D. T. Keating and A. N. Goland, J. Appl. Phys. **39**, 6018 (1968).

[21] L. D. Eshelby, Proc. Roy. Soc. (London) **A241**, 376 (1957).

[22] This result is due to linear continuum theory. If nonlinear terms are included there is always a volume change, which is of the order of one unit volume per atomic length of the dislocation. H. Stehle and A. Seeger, Z. Physik **146**, 217 (1956).

from (31)

$$\langle(\mathbf{s}^m)^2\rangle = c(1-c)\sum_n (\mathbf{t}^{m-n})^2 + c\sum_{n\neq n'} g(n-n')\mathbf{t}^{m-n}\cdot\mathbf{t}^{m-n'}. \quad (33)$$

To evaluate the sums, we use the continuum expression (20) for $\mathbf{t}(\mathbf{r})$ and replace the sums by integrations. The first term, giving the only contribution for a random distribution, diverges for small r and has to be cut off at the ion radius d_c. This indicates already that the continuum expression (20) gives only a rough estimate of this term, which depends sensitively on the displacements of the atoms in the nearest-neighbor shell around the defect.[6,14] The second term in (33) describes the clustering effect and is, for large clusters, not very sensitive to the values at small r,r' or $|\mathbf{r}-\mathbf{r}'|$. Therefore, the continuum approximation is good for this term. Integration by parts gives us, considering that $\mathbf{t}=-A\nabla_r r^{-1}$ and $\nabla_r^2 r^{-1} = -4\pi\delta(\mathbf{r})$,

$$\langle(\mathbf{s}^m)^2\rangle \simeq c\frac{4\pi A^2}{V_c}\frac{1}{d_c} + c\frac{4\pi A^2}{V_c}\frac{n_{\rm cl}}{R_{\rm cl}},$$

with

$$\frac{1}{R_{\rm cl}} = \frac{1}{n_{\rm cl}}\int \frac{d\mathbf{r}}{V_c}\frac{1}{r} g(\mathbf{r}). \quad (34)$$

Due to the clustering, $\langle\mathbf{s}^2\rangle$ (hence L_h and $\mu^{\rm PE}$) increases essentially by a factor $n_{\rm cl}\cdot d_c/R_{\rm cl}$, where $R_{\rm cl}$ is an average cluster radius. For spherical clusters with a uniform defect density ρ_0 inside the cluster and a cluster radius R_0, we have $R_{\rm cl}=\frac{5}{6}R_0$ and $n_{\rm cl}=\rho_0(4\pi/3)(R_0^3/V_c)$. As an example, we consider clusters in Cu and assume $\rho_0=10^{-2}$ and $R_0=50$ Å, and, thus, $n_{\rm cl}=445$. For these values, we get an increase of the photoelectric absorption by a factor of 11.

The foregoing discussion is based on formula (28) for the Debye-Waller factor. For the case of statistically distributed point defects,[6,23] it has been shown that this formula is only a poor approximation for $L_{\rm h}$. To discuss its validity in the case of clusters, we assume the defects in the cluster are independently distributed, giving rise to a cluster-displacement field

$$\mathbf{t}_{\rm cl}(\mathbf{r}) = \int (d\mathbf{r}'/V_c)\rho_0(\mathbf{r}')A(\mathbf{r}-\mathbf{r}')/|\mathbf{r}-\mathbf{r}'|^3 = t_{\rm cl}(r)\mathbf{r}/r, \quad (35)$$

with

$$t_{\rm cl}(r) = \frac{4\pi A}{V_c}\frac{1}{r^2}\int_0^r r'^2 dr'\rho_0(r').$$

The $\rho_0(r')$ is the defect density within the spherical clusters. If the cluster centers are statistically distributed, we get for $L_{\rm h}$,[6,23] in the same way as it will be derived in (39) and (40) for loops ($c_{\rm cl}$=concentration of clusters per lattice site),

$$L_{\rm h} = c_{\rm cl}\int \frac{d\mathbf{r}}{V_c}[1-\cos\mathbf{h}\cdot\mathbf{t}_{\rm cl}(\mathbf{r})] = c_{\rm cl}\int \frac{4\pi r^2 dr}{V_c}\left(1-\frac{\sin h t_{\rm cl}(r)}{h t_{\rm cl}(r)}\right). \quad (36)$$

Due to spherical symmetry of the clusters and the isotropic approximation (20), $L_{\rm h}$ depends only on $h=|\mathbf{h}|$. It is convenient to expand $L_{\rm h}$ into powers of h^2:

$$L_{\rm h} = L_{\rm h}^{(0)} + L_{\rm h}^{(1)} + \cdots + L_{\rm h}^{(n)} + \cdots, \quad (37)$$

with

$$L_{\rm h}^{(n)} = c_{\rm cl}\int_0^\infty \frac{4\pi r^2 dr}{V_c}\frac{(-1)^n}{(2n+3)!}[h t_{\rm cl}(r)]^{2n+2}.$$

For clusters with a constant density $\rho_0(r)=\rho_0$ and radius R_0, we get for this, using Eq. (35),

$$L_{\rm h}^{(n)} = c_{\rm cl}\frac{4\pi R_0^3}{V_c}\frac{(-1)^n}{(2n+3)!}\left(\frac{1}{2n+5}+\frac{1}{4n+1}\right)\times\left(\rho_0\frac{4\pi A}{V_c}\frac{hR_0}{3}\right)^{2n+2}. \quad (38)$$

The first term $L_{\rm h}^0$ gives the same result as the second term in Eq. (34). Because (38) is an expansion into powers of $(\rho_0 A h R_0)$, the higher-order terms are more important for very large or dense clusters or for higher-order reflections. Because our cluster model is reasonable only for small concentration ρ_0, the higher-order terms are generally small, if R_0 or h is not too large. For example, for interstitials in Cu we have $4\pi A/V_c\approx 1$ and if $\rho_0=10^{-2}$, the condition $L_{\rm h}^{(1)}\leq\frac{1}{10}L_{\rm h}^{(0)}$ holds for $R_0\leq 260$ Å ($n_{\rm cl}\leq 6.4\times10^4$) for a $\langle111\rangle$ reflection, and $R_0\leq 87$ Å ($n_{\rm cl}\leq 2.4\times10^3$) for a $\langle333\rangle$ reflection. In Sec. V, we will see that for the dislocation loop model, representing a very dense cluster, the higher-order terms are very important.

For the general anisotropic displacement field (19), the above discussion based on formula (28) is only slightly changed. One ends up with the same expression (34) but with a somewhat different constant A. Significantly different are only the higher-order terms $L_{\rm h}^{(n)}$, e.g., $L_{\rm h}^{(1)}$ depends not only on h^4, but also on $\sum_i h_i^4$.

V. DEBYE-WALLER FACTOR FOR LOOP MODEL

For the case that the point defects form dislocation loops, we have $\mathbf{s}^m=\sum_n p_L^n\mathbf{t}^{m-n}$, where $\mathbf{t}^{m-n}$ is the displacement of atom m due to a loop with center at n and p_L^n is a random number (12) indicating whether there is such a loop with its center at n or not. Considering that $(p_L^n)^2=p_L^n$, $\langle p_L^n\rangle=C_L$ and for $n\neq n'$, $\langle p_L^n p_L^{n'}\rangle = C_L^2$ (etc., C_L=loop concentration per lattice site), if

[23] M. A. Krivogalz, Fiz. Metal Metalloved. 7, 250 (1959) [English transl.: Phys. Metals Metallog. (USSR) 7, 11 (1959)].

the loop centers are statistically distributed, we get

$$\langle\exp(i\mathbf{h}\cdot\mathbf{s}^m)\rangle = \langle\prod_n [1+p_L{}^n(\exp(i\mathbf{h}\cdot\mathbf{t}^{m-n}-1))]\rangle = \prod_n [1+C_L(\exp(i\mathbf{h}\cdot\mathbf{t}^{m-n}-1))] = \exp[-C_L\sum_n (1-\cos\mathbf{h}\cdot\mathbf{t}^{m-n})]. \quad (39)$$

In the last line, we assumed that the loop concentration $C_L \ll 1$, which is, of course, always the case. Using the continuum expression (24) for $\mathbf{t}$ and considering that for a loop with an average radius R_0, $\mathbf{t}(\mathbf{r})=\boldsymbol{\tau}(\mathbf{r}/R_0)$ depends only on the reduced coordinate $\mathbf{r}=\mathbf{r}/R_0$, the Debye-Waller factor is given by

$$L_\mathbf{h} = C_L\int \frac{d\mathbf{r}}{V_c}[1-\cos\mathbf{h}\cdot\mathbf{t}(\mathbf{r})] = C_L\frac{R_0{}^3}{V_c}\int d\tilde{\mathbf{r}}[1-\cos\mathbf{h}\cdot\boldsymbol{\tau}(\tilde{\mathbf{r}})] = C_L R_0{}^3/V_c\, l(\mathbf{h},\mathbf{b}). \quad (40)$$

Therefore, $L_\mathbf{h}$ is directly proportional to $R_0{}^3$ and the remaining integral $l(\mathbf{h},\mathbf{b})$ no longer depends on the size, but only on the shape and Burgers vector of the loop. Taking into account that the number n_L of point defects forming a loop is $n_L=\pi R_0{}^2\cdot b/V_c$, we have $L_\mathbf{h}\simeq C_L R_0{}^3/V_c \sim cR_0/b$ with $C_L\cdot n_L=c$. Thus, if point defects form a dislocation loop, the Debye-Waller factor and μ^{PE} increase by a factor of R_0/b.

The remaining integral $l(\mathbf{h},\mathbf{b})$ in (40) cannot be performed in general due to the complicated (and usually unknown) displacement field $\boldsymbol{\tau}(\mathbf{r})$. Following Krivoglaz,[24] we replace $\mathbf{t}(\mathbf{r})$ by the asymptotic expression (25), an approximation which is allowed for $hb\gg 1$, but which seems to be surprisingly good also for moderate values of hb.[24] Setting $\mathbf{t}(\mathbf{r})=\frac{1}{2}b(R_0/r)^2\boldsymbol{\tau}(\Omega)$, one gets with $\mathbf{h}=h\mathbf{h}^0$ and $\mathbf{b}=b\mathbf{b}^0$ ($\mathbf{h}^{0^2}=1=\mathbf{h}^{0^2}$)

$$l(\mathbf{h},\mathbf{b}) = (hb)^{3/2}\eta(\mathbf{h}^0,\mathbf{b}^0)$$

with

$$\eta(\mathbf{h}^0,\mathbf{b}^0) = \frac{\sqrt{\pi}}{6}\int d\Omega[\mathbf{h}^0\cdot\boldsymbol{\tau}(\Omega,\mathbf{b}^0)]^{3/2}. \quad (41)$$

It should be noted that the $h^{3/2}$ dependence resulting from the large displacement field ($\sim b$) of the dense dislocation loop is much less than the h^2 behavior of loose clusters (28).

Considering that we may have loops on all {111} planes and/or all {110} planes, we must average (41) over all the cubic equivalent Burgers vectors $\mathbf{b}_i$ ($i=1,\ldots,z$),

$$\bar{\eta}(\mathbf{h}^0,\mathbf{b}^0) = \frac{1}{z}\sum_{i=1}^{z}\eta(\mathbf{h}^0,\mathbf{b}_i{}^0) = \frac{1}{z'}\sum_{j=1}^{z'}\eta(\mathbf{h}_j{}^0,\mathbf{b}^0). \quad (42)$$

Here, we can replace the average over $\mathbf{b}_i{}^0$ by a similar average over all cubic equivalent $\mathbf{h}_j{}^0$, $j=1,\ldots,z'$. Due to this average, the result is not very sensitive to the direction of $\mathbf{h}^0$. For example, the integral (41) can be evaluated approximately by Houston's formula.[25,26] In the isotropic case, using the asymptotic field (26) with $\nu=\frac{1}{3}$, one gets the following values for $\bar{\eta}(\mathbf{h}^0,\mathbf{b}^0)$: $\mathbf{h}^0=\langle 100\rangle$, $\langle 110\rangle$, $\langle 111\rangle$; and for $\mathbf{b}^0=\langle 100\rangle$; $\bar{\eta}=0.460$, 0.495, 0.515; $\mathbf{b}^0=\langle 110\rangle$: $\bar{\eta}=0.495$, 0.495, 0.485; $\mathbf{b}^0=\langle 111\rangle$: $\bar{\eta}=0.515$, 0.485, 0.495. Therefore in a reasonable approximation $L_\mathbf{h}$, is given by

$$L_\mathbf{h}\approx C_L(R_0{}^3/V_c)\tfrac{1}{2}(hb)^{3/2}. \quad (43)$$

VI. DIFFUSE SCATTERING ABSORPTION FOR CLUSTER MODEL

To calculate the diffuse scattering absorption, we have to start with the imaginary potential coefficients $I_{\mathbf{h},\mathbf{h}'}{}^{(2)ss'}$ (17). Due to the low defect concentrations of our clusters, the total displacements $\mathbf{s}^m$ and $\mathbf{s}^{m'}$ in (18) are normally small. Therefore, we can expand $S_{\mathbf{h}\mathbf{h}'}(\mathbf{k}-\mathbf{k}')$ into powers of $\mathbf{s}^m$, taking only the lowest term into account.

$$S_{\mathbf{h}\mathbf{h}'}(\mathbf{k}-\mathbf{k}') = \sum_{m-m'}\exp i\mathbf{q}\cdot(\bar{\mathbf{R}}^m-\bar{\mathbf{R}}^{m'}) \times\langle[(\mathbf{k}+\mathbf{h}-\mathbf{k}')\cdot\mathbf{s}^m][(\mathbf{k}+\mathbf{h}'-\mathbf{k}')\cdot\mathbf{s}^{m'}]\rangle. \quad (44)$$

Quite analogously to (33), we get for the displacement correlation $\langle s_i{}^m s_j{}^{m'}\rangle$ in the cluster model ($c\ll 1$),

$$\langle s_i{}^m s_j{}^{m'}\rangle = c\sum_n t_i{}^{m-n}t_j{}^{m'-n} + c\sum_{n\neq n'} t_i{}^{m-n}g(n-n')t_j{}^{m'-n'}. \quad (45)$$

The first term gives the only contribution for a random distribution of point defects, whereas the second term is due to the clustering. The importance of the clustering term depends very much on the distance $|\bar{\mathbf{R}}^m-\bar{\mathbf{R}}^{m'}|$. For small $|\bar{\mathbf{R}}^m-\bar{\mathbf{R}}^{m'}|$, e.g., for $m=m'$, the average $\langle(\mathbf{s}^m)^2\rangle$ increases by a factor $n_{\mathrm{cl}}d_c/R_{\mathrm{cl}}$ due to clustering [Eq. (34)]. For large $|\bar{\mathbf{R}}^m-\bar{\mathbf{R}}^{m'}|$, on the other hand, the average (34) increases even by a factor n_{cl}, because then $t_j{}^{m'-n'}$ in (45) can be replaced by $t_j{}^{m'-n}$ and the sum over n' gives a factor n_{cl}. Inserting (45) into (44), we get

$$S_{\mathbf{h},\mathbf{h}'}(\mathbf{k}-\mathbf{k}') = c[(\mathbf{k}+\mathbf{h}-\mathbf{k}')\cdot\tilde{\mathbf{t}}(\mathbf{q})] \times[(\mathbf{k}+\mathbf{h}'-\mathbf{k}')\cdot\tilde{\mathbf{t}}^*(\mathbf{q})][1+\tilde{g}(q)], \quad (46)$$

[24] For $h=b=[111]$, and for isotropy, we have checked this by numerical integration. The result was $\approx 15\%$ higher than given by Eqs. (41) and (43)].

[25] M. A. Krivolgaz and K. P. Ryaboshka, Fiz. Metal. Metalloved. **16**, 641 (1963) [English transl.: Phys. Metals Metallog. (USSR) **17**, 1 (1953).

[26] D. D. Betts, A. B. Bhatia, and M. Wyman, Phys. Rev. **104**, 37 (1956).

with

$$\tilde{g}(\mathbf{q})=\sum_{n\neq 0} e^{i\mathbf{q}\cdot\bar{\mathbf{R}}^n}g(n)\simeq\int\frac{d\mathbf{r}}{V_c}e^{i\mathbf{q}\cdot\mathbf{r}}g(r)$$

and

$$\tilde{\mathbf{t}}(\mathbf{q})=\sum_{m} e^{i\mathbf{q}\cdot\bar{\mathbf{R}}^m}\mathbf{t}^m\simeq\int\frac{d\mathbf{r}}{V_c}e^{i\mathbf{q}\cdot\mathbf{r}}\mathbf{t}(\mathbf{r}).$$

The replacement of the sums by integrals is only valid for q values small compared with a reciprocal-lattice vector, which are the most important ones in the following. The Fourier transform $\tilde{g}(\mathbf{q})\simeq n_{\mathrm{cl}}$ for small values of $q\leq 1/R_{\mathrm{cl}}$ due to the normalization of $g(r)$. Therefore, $S_{\mathbf{hh}'}(\mathbf{k}-\mathbf{k}')$ increases due to the clustering for small q values by a factor of n_{cl} which corresponds just to the increase of the long-range correlations (45). But there is no essential increase of $S_{\mathbf{hh}'}(\mathbf{k}-\mathbf{k}')$ for larger q values for which $\tilde{g}(\mathbf{q})$ decreases very rapidly.

In the isotropic case (20), we get, from (46),

$$S_{\mathbf{hh}'}(\mathbf{k}-\mathbf{k}')=c(4\pi A/V_c)^2\{[(\mathbf{k}+\mathbf{h}-\mathbf{k}')\cdot\mathbf{q}]\times[(\mathbf{k}+\mathbf{h}'-\mathbf{k}')\cdot\mathbf{q}]/q^4\}[1+\tilde{g}(q)]. \quad (47)$$

In order to see whether the coefficients $I_{\mathbf{hh}'}{}^{(2)}$ and μ^{DS} are affected by the defect clustering, we have to look for such small q regions in the $\mathbf{k}'$ space which lie near to the integration surface $\mathbf{k}'^2=k^2$ in (17), i.e., the Ewald sphere.

In the one-beam case, where no Bragg reflections are excited, we have, in general, only small q values with $\tilde{g}(\mathbf{q})\simeq n_{\mathrm{cl}}$ in the forward direction $\mathbf{k}'=\mathbf{k}+\mathbf{q}\simeq\mathbf{k}$, corresponding to the well-known small-angle scattering. The coefficient $I_{00}{}^{ss}$, the only one of interest in the one-beam case, can be calculated from (47) for $\mathbf{k}'=\mathbf{k}+q$:

$$S_{00}(\mathbf{k}-\mathbf{k}')=c(4\pi A/V_c)^2[1+\tilde{g}(q)]. \quad (48)$$

[Equation (48) represents only the small-angle scattering at the lattice dilatation introduced by the clusters. The "direct" scattering at the defect atoms has been neglected from the beginning.] Since only in a very small area $\sim 1/R_{\mathrm{cl}}{}^2$ in the forward direction is $\tilde{g}(\mathbf{q})\approx n_{\mathrm{cl}}$, we get, for the contribution of the small-angle scattering to the diffuse scattering absorption, $\mu^{\mathrm{DS}}\sim n_{\mathrm{cl}}\cdot(\lambda/R_{\mathrm{cl}})^2$. Therefore, in the one-beam case, μ^{DS} is not very sensitive to clustering and not important compared with μ^{PE}.

In the two-beam case, where two plane waves $\mathbf{k}$ and $\mathbf{k}+\mathbf{h}$ are strongly excited, we get, besides the small angle region, an additional region with small q values near the Bragg reflection $\mathbf{k}'=\mathbf{k}+\mathbf{h}+\mathbf{q}\simeq\mathbf{k}+\mathbf{h}$. For these $\mathbf{k}'$ values, S_{00} is given by

$$S_{00}(\mathbf{k}-\mathbf{k}')=c(4\pi A/V_c)^2[(\mathbf{h}+\mathbf{q})\cdot\mathbf{q}/q^2]^2[1+\tilde{g}(q)]. \quad (49)$$

For randomly distributed point defects $\tilde{g}=0$. This represents the Huang scattering[27] of the plane wave $\mathbf{k}$ very near to the Bragg reflection and varies as $(h/q)^2$ for small q's. Due to the clustering, this Huang scattering is now enhanced by a factor of n_{cl} for $q\leq 1/R_{\mathrm{cl}}$. Compared with the small-angle scattering (48), it is larger by a factor of $(hR_{\mathrm{cl}})^2$; for $\mathbf{h}=[111]$ in Cu and $R_{\mathrm{cl}}=30$ Å, this factor is 10^4. Therefore, μ^{DS} will be very sensitive to clustering in the two-beam case.

For $S_{\mathbf{hh}}(\mathbf{k}-\mathbf{k}')$, we get the same expression as (49) for S_{00}, but for $\mathbf{k}'$ values $\mathbf{k}'=\mathbf{k}+\mathbf{q}\simeq\mathbf{k}$, representing the Huang scattering of the plane wave $\mathbf{k}+\mathbf{h}$ near to the Bragg reflection $\mathbf{k}'\approx\mathbf{k}$. The mixed coefficients $S_{0\mathbf{h}}=S_{\mathbf{h}0}$ represent the interference scattering of the plane waves $\mathbf{k}$ and $\mathbf{k}+\mathbf{h}$ and are for small q's proportional to $(h/q)n_{\mathrm{cl}}$. Therefore, they are a factor $(hR_{\mathrm{cl}})^{-1}$ smaller than S_{00} and $S_{\mathbf{hh}}$ and are neglected in the following. To get the coefficients $I_{00}{}^{ss}$ and $I_{\mathbf{hh}}{}^{ss}$, we can replace in (17), for $\mathbf{k}'\simeq\mathbf{k}+\mathbf{h}$, $f_{\mathbf{k}-\mathbf{k}'}$ by $f_{\mathbf{h}}'$ and, moreover, the integration surface $\mathbf{k}'^2=\mathbf{k}^2$ by the tangential plane at the Bragg reflection. If the Bragg condition is exactly fulfilled $[\mathbf{k}^2=(\mathbf{k}+\mathbf{h})^2]$, the integrals $I_{00}{}^{ss}$ and $I_{\mathbf{hh}}{}^{ss}$ diverge due to the $1/q^2$ behavior of S_{00} and $S_{\mathbf{hh}}$. This divergence is due to the substitution of the lattice Green's function $G(\mathbf{r},\mathbf{r}')$ (5) by the free Green's function, which does not apply for $\mathbf{k}'$ waves scattered directly into the Bragg condition. This can be taken into account by cutting off the q integration at $q_c=(k^2\chi_h e^{-M_h})/h$,[3,4] which is of the order of the reciprocal extinction length. Then we have

$$I_{00}=-\frac{2\pi}{V_c}\left(\frac{e^2}{mc^2}\right)^2P^2f_{\mathbf{h}}'^2c\left(\frac{4\pi A}{V_c}\right)^2\frac{1}{k}\times\int_{q\geq q_c}qdq\frac{h^2}{q^2}g(q)\int_{\substack{0\\ \boldsymbol{\kappa}\perp\mathbf{k}+\mathbf{h}}}^{2\pi}\frac{d\phi}{2\pi}(\mathbf{h}^0\cdot\boldsymbol{\kappa})^2, \quad (50)$$

with $\mathbf{h}^0=\mathbf{h}/h$, $\boldsymbol{\kappa}=\mathbf{q}/q$, and $P=1$ or $\cos 2\theta_B$ for σ or π polarization, where ϕ is the azimuth in the tangential plane $\boldsymbol{\kappa}\perp\mathbf{k}+\mathbf{h}$. For $I_{\mathbf{hh}}$, one gets the same integral but over the tangential plane $\boldsymbol{\kappa}\perp\mathbf{k}$. Due to the isotropic expression (20) for $\mathbf{t}(\mathbf{r})$, both integrals are equal $I_{00}{}^{ss}=I_{\mathbf{hh}}{}^{ss}$. Therefore, we get for the diffuse scattering absorption, considering that $I_{0\mathbf{h}}{}^{ss}\simeq 0\simeq I_{\mathbf{h}0}{}^{ss}$,

$$\mu^{\mathrm{DS}}=-\frac{1}{k}I_{00}=\frac{\pi}{V_c}\left(\frac{e^2}{mc^2}Pf_{\mathbf{h}}'\right)^2\cdot\left(\frac{h}{k}\right)^2c\left(\frac{4\pi A}{V_c}\right)^2\times\cos 2\theta_B\ln\frac{q_{\mathrm{cl}}}{q_c}. \quad (51)$$

Here, $q_{\mathrm{cl}}=1/R_{\mathrm{cl}}$ is the cutoff at large q values and R_{cl} is an effective cluster radius. Due to the log dependence, the actual values of q_c and q_{cl} are not very important. Hence, μ^{DS} varies as $f_{\mathbf{h}}'^2\cdot\mathbf{h}^2$ as long as $\cos\theta_B\approx 1$, and is proportional to λ^2. It is important to realize that, since $I_{0\mathbf{h}}\simeq 0$ and $I_{00}=I_{\mathbf{hh}}$, μ^{DS} has the same value on both branches of the dispersion surface, quite in contrast to the photoelectric absorption μ^{PE} (15). If the Bragg condition is not exactly fulfilled, the minimum q value is

[27] K. Huang, Proc. Roy. Soc. (London) **A190**, 102 (1947).

given by the excitation error $q_0=\big||\mathbf{k}+\mathbf{h}|-\mathbf{k}\big|$. Also, in this case, we can have a very intense diffuse scattering. And as long as $q_0\ll 1/R_{\rm cl}$, we can in Eq. (51) for $\mu^{\rm DS}$ simply replace q_c by $(q_c^2+q_0^2)^{1/2}$. Therefore, $\mu^{\rm DS}$ is practically constant over the whole Bragg reflection and goes only slowly to zero for relatively large q_0 values. This means that also in the one-beam case the normal absorption increases considerably due to the diffuse scattering, if one *approaches the Bragg reflection*, long before the second beam is strongly excited. This behavior of $\mu^{\rm DS}$ (51) and $\mu^{\rm PE}$ (15) is plotted in Fig. 1 as a function of W [Eq. (10)]. Due to the Huang scattering from the clusters, $\mu^{\rm total}=\mu^{\rm PE}+\mu^{\rm DS}$ has "shoulders" for large W values.

In a similar way, in the two-beam case, one can also get an additional increase in the absorption, if one approaches a multiple-beam case, namely, due to the strong scattering near new reflections. Here, the additional absorption is in general different for the two branches, as can be derived easily from (17) and (47).

The effects of anisotropy can be calculated by using the perturbation expression (21). Compared with the isotropic expression (50), only the integral over ϕ is now different if we define A by $4\pi A=P_0/(c_{12}^{\,v}+2c_{44}^{\,v})$. For I_{00}, this integral, called j_{00}, is

$$j_{00}=\int_{\substack{0\\ \kappa\perp\mathbf{k}+\mathbf{h}}}^{2\pi}\frac{d\phi}{2\pi}|\mathbf{h}^0\cdot\boldsymbol{\gamma}(\boldsymbol{\kappa})|^2$$

with

$$\gamma_i(\boldsymbol{\kappa})=\kappa_i+(d/c_{44}^{\,v})\Big[-\kappa_i^3+\beta^v\kappa_i\sum_m\kappa_m^4+\tfrac{3}{5}(1-\beta^v)\kappa_i\Big]. \quad (52)$$

Taking only the linear term in $d/c_{44}^{\,v}$ into account, we get

$$\begin{aligned}j_{00}=&\tfrac{1}{2}[1-(\mathbf{h}^0\cdot\mathbf{e})^2]\\&+2(d/c_{44}^{\,v})\{[-(6/80)+(1/80)\beta][1-(\mathbf{h}^0\cdot\mathbf{e})^2]\\&+(\tfrac{3}{8}-\tfrac{1}{4}\beta)\sum_i h_i^{0\,2}e_i^2+(-\tfrac{3}{8}+\tfrac{1}{2}\beta)\mathbf{h}^0\cdot\mathbf{e}\sum_i h_i^0e_i^3\\&+\tfrac{1}{16}\beta\sum_m e_m^4[1-5(\mathbf{h}^0\cdot\mathbf{e})^2]\},\end{aligned} \quad (53)$$

with

$$\mathbf{e}=(\mathbf{k}+\mathbf{h})/|\mathbf{k}+\mathbf{h}|\,,$$

which is equal to $\frac{1}{2}\cos^2\theta_B$ for $d\equiv 0$. For $I_{\rm hh}$, we get the same expression but with $\mathbf{e}=\mathbf{k}/k$. Due to the anisotropy, $\mu^{\rm DS}$ depends explicitly on the direction of $\mathbf{k}$ with respect to the cubic axes as well as its direction with respect to $\mathbf{h}$, i.e., the Bragg angle θ_B. Therefore, $\mu^{\rm DS}$ changes, if, for a fixed Bragg angle θ_B, $\mathbf{k}$ is rotated around the direction of $\mathbf{h}$ which is indicated in Fig. 2. For $\theta_B\simeq 0$, we have plotted j_{00} for Cu as a function of the rotation angle ϕ for $\mathbf{h}^0=[100]$ (fourfold symmetry) and $\mathbf{h}^0=[110]$ (twofold symmetry). In the isotropic case, both curves would be identical and equal to $\frac{1}{2}$ (dotted line).

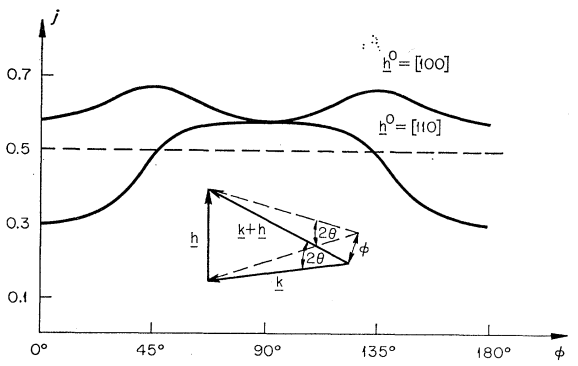

FIG. 2. Dependence of the diffuse scattering absorption $\mu^{\rm DS}$ on the rotation angle ϕ.

A second consequence of the anisotropy is that the coefficients I_{00} and $I_{\rm hh}$ are in general different because of the different directions of the tangential planes with respect to the cubic axes. Execptions are symmetrical reflections of the type $\langle 100\rangle$ and $\langle 110\rangle$ and, furthermore, large $\mathbf{k}$ values for which $\mathbf{k}+\mathbf{h}\simeq\mathbf{k}(\theta_B\approx 0)$. According to (9), we get in the general case for the absorption

$$\begin{aligned}\mu^{\rm DS}=-(1/k)\big[&\tfrac{1}{2}(I_{00}^{\,ss}+I_{\rm hh}^{\,ss})\\&\mp W(1+W^2)^{-1/2}\,\tfrac{1}{2}(I_{00}^{\,ss}-I_{\rm hh}^{\,ss})\big].\end{aligned} \quad (54)$$

$\mu^{\rm DS}$ varies from $-(1/k)I_{00}^{\,ss}$ on the one side of the Bragg reflection to $-(1/k)I_{\rm hh}^{\,ss}$ on the other side and just opposite on the two branches of the dispersion surface, as illustrated in Fig. 3. It causes an asymmetry of the total absorption $\mu^{\rm total}=\mu^{\rm PE}+\mu^{\rm DS}$.

According to (51), $\mu^{\rm DS}$ vanishes in the isotropic case for $\theta_B\to 90°$. In the anisotropic case, it vanishes only for symmetrical reflections, but according to (53), always if we consider only linear terms in d.

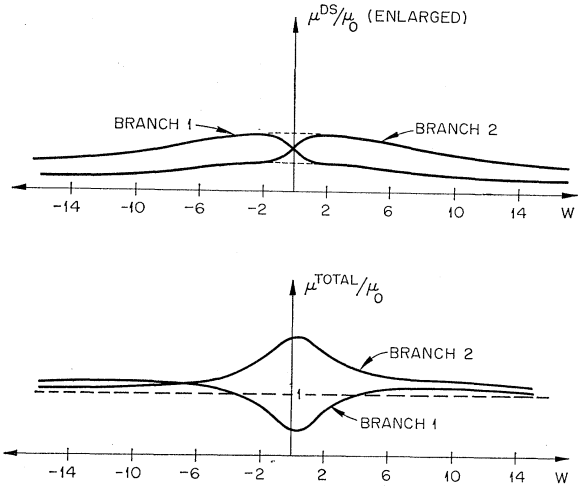

FIG. 3. Asymmetry of $\mu^{\rm DS}$ and $\mu^{\rm total}$ for nonsymmetrical reflections.

VII. DIFFUSE SCATTERING ABSORPTION FOR LOOP MODEL

Assuming that the loop centers are independently distributed and considering that the loop concentration C_L per lattice site is always small, we get for $S_{\mathbf{hh}'}$

$$\begin{aligned}S_{\mathbf{hh}'}(\mathbf{k}-\mathbf{k}')&=\sum_{m-m'}\exp i\mathbf{q}\cdot(\bar{\mathbf{R}}^m-\bar{\mathbf{R}}^{m'})\\&\times[\exp C_L\sum_n(\exp[-i(\mathbf{k}+\mathbf{h}-\mathbf{k}')\cdot\mathbf{t}^{m-n}\\&\qquad+i(\mathbf{k}+\mathbf{h}'-\mathbf{k}')\cdot\mathbf{t}^{m'-n}]-1)\\&-\exp C_L\sum_n(\exp-i(\mathbf{k}+\mathbf{h}-\mathbf{k}')\cdot\mathbf{t}^{m-n}-1)\\&\times\exp C_L\sum_n(\exp i(\mathbf{k}+\mathbf{h}'-\mathbf{k}')\cdot\mathbf{t}^{m'-n}-1)].\end{aligned}\tag{55}$$

This can be derived in the same way as the expression (39) for the Debye-Waller factor if one replaces $\mathbf{h}\cdot\mathbf{s}^m$ by $-(\mathbf{k}+\mathbf{h}-\mathbf{k}')\cdot\mathbf{s}^m+(\mathbf{k}+\mathbf{h}'-\mathbf{k}')\cdot\mathbf{s}^{m'}$. If the exponents or the Debye-Waller factors (43) are small, we can expand linearly in the concentration C_L

$$S_{\mathbf{hh}'}(\mathbf{k}-\mathbf{k}')=C_LG_{\mathbf{k}+\mathbf{h}-\mathbf{k}'}(\mathbf{q})G_{\mathbf{k}+\mathbf{h}'-\mathbf{k}'}{}^*(\mathbf{q}),\tag{56}$$

with

$$\begin{aligned}G_{\mathbf{k}+\mathbf{h}-\mathbf{h}'}(\mathbf{q})&=\sum_n\exp i\mathbf{q}\cdot\bar{\mathbf{R}}^m(1-\exp-i(\mathbf{k}+\mathbf{h}-\mathbf{k}')\cdot\mathbf{t}^m)\\&\simeq\int\frac{d\mathbf{r}}{V_c}\exp i\mathbf{q}\cdot\mathbf{R}(1-\exp-i(\mathbf{k}+\mathbf{h}-\mathbf{k}')\cdot\mathbf{t}(\mathbf{r})).\end{aligned}$$

Due to the slow variation of $t(\bar{\mathbf{R}}^m)$ for a dislocation loop, the sum can be replaced by the integral. Because $\mathbf{t}(\mathbf{r})$ depends only on the reduced coordinate $\mathbf{r}/R_0$, $G(\mathbf{q})$ depends only on $\mathbf{q}\cdot R_0$. For $qR_0\leq1$, the most important contribution to the integral comes from the large r values, for which we can replace $\mathbf{t}(\mathbf{r})$ by the asymptotic field (25) or (26) and for which we can expand (56) linearly in $\mathbf{t}(\mathbf{r})$. Then we get, for $qR_0\ll1$,

$$\begin{aligned}S_{\mathbf{hh}'}(\mathbf{k}-\mathbf{k}')&\\&=C_L(\mathbf{k}+\mathbf{h}-\mathbf{k}')\cdot\mathbf{t}_0(\mathbf{q})(\mathbf{k}+\mathbf{h}'-\mathbf{k}')\cdot\mathbf{t}_0{}^*(\mathbf{q}),\end{aligned}\tag{57}$$

where, in the isotropic case, $\mathbf{t}_0(\mathbf{q})$ is given by ($\mathbf{b}^0=\mathbf{b}/|b|$, $\boldsymbol{\kappa}=\mathbf{q}/q$),

$$\begin{aligned}\mathbf{t}_0(\mathbf{q})&=\frac{i}{q}\frac{bF}{V_c}\boldsymbol{\eta}(\boldsymbol{\kappa},\mathbf{b}^0)\\&=\frac{i}{q}\frac{bF}{V_c}\left[2\mathbf{b}^0(\mathbf{b}^0\cdot\boldsymbol{\kappa})+\frac{\nu}{1-\nu}\boldsymbol{\kappa}-\frac{1}{1-\nu}(\mathbf{b}^0\cdot\boldsymbol{\kappa})^2\boldsymbol{\kappa}\right].\end{aligned}\tag{58}$$

For larger q values, $qR_0\geq1$, the approximations leading to (57) are no longer allowed and $S_{\mathbf{hh}'}$ vanishes much more rapidly than $1/q^2$, as in (57). Considering that the number n_L of point defects contained in one dislocation loop is $n_L=bF/V_c$, we see that $S_{\mathbf{hh}'}$ is essentially proportional to $c_Ln_L{}^2/q^2=cn_L/q^2$, which is equivalent to the results for the cluster model (46). The reason for this is, of course, that the diffuse scattering for small q is determined by the asymptotic displacement fields, which are in both models proportional to n_{cl}/r^2. Hence, in the one-beam case, we get no strong absorption in agreement with the cluster model because the total small-angle intensity is not very large. In the two-beam case, the Fourier coefficient I_{00} is essentially determined by the extremely strong diffuse scattering of the plane wave $\mathbf{k}$ near the Bragg reflection $\mathbf{k}+\mathbf{h}$. From (57), we get for this

$$\begin{aligned}I_{00}{}^{ss}=-\frac{2\pi}{V_c}&[(e^2/mc^2)Pf_{\mathbf{h}}{}']^2C_L(bF/V_c)^2(h^2/k)\\&\times[\ln(\kappa_0/q_c)]j[(\mathbf{k}+\mathbf{h})/|\mathbf{k}+\mathbf{h}|].\end{aligned}\tag{59}$$

Here, j is an angle integral in the tangential plane $\boldsymbol{\kappa}\perp\mathbf{k}+\mathbf{h}$.

$$j\left(\frac{\mathbf{k}+\mathbf{h}}{|\mathbf{k}+\mathbf{h}|}\right)=\frac{1}{z}\sum_{i=1}^{z}\int_{\boldsymbol{\kappa}\perp\mathbf{k}+\mathbf{h}}^{2\pi}\frac{d\phi}{2\pi}|\mathbf{h}^0\cdot\boldsymbol{\eta}(\boldsymbol{\kappa},\mathbf{b}_i{}^0)|^2.\tag{60}$$

Furthermore, we have summed in (60) over all those dislocation loops $\mathbf{b}_i{}^0$ ($i=1,\ldots,z$) which are equivalent due to cubic symmetry and occur always with the same probability (e.g., all $\langle111\rangle$ directions, $z=8$). For $I_{\mathbf{hh}}$, we get the same expression as (59) but with $j=j(\mathbf{k}/k)$, corresponding to the diffuse scattering of the plane wave $\mathbf{k}+\mathbf{h}$ into the forward direction $\mathbf{k}'\simeq\mathbf{k}$. The coefficients $I_{0\mathbf{h}}$ and $I_{\mathbf{h}0}$ corresponding to the interference scattering between $\mathbf{k}$ and $\mathbf{k}+\mathbf{h}$ is an order of magnitude smaller than I_{00} and $I_{\mathbf{hh}}$ and will be neglected.

Introducing $\boldsymbol{\eta}(\boldsymbol{\kappa},\mathbf{b}^0)$ from (58) into (60), we get for $j(\mathbf{e})$

$$\begin{aligned}j(\mathbf{e})=4\alpha+\tfrac{1}{2}&[1-(\mathbf{h}^0\cdot\mathbf{e})^2]\left(\frac{\nu(2-\nu)}{3(1-\nu)^2}-\frac{1+8\nu-8\nu^2}{(1-\nu)^2}\right)\\&+\beta\Big\{2(1-\sum_i h_i{}^{02}e_i{}^2)\\&-\frac{3}{2(1-\nu)}[1-(\mathbf{h}^0\cdot\mathbf{e})^2-\sum_i h_i{}^{02}e_i{}^2+\mathbf{h}^0\cdot\mathbf{e}\sum_i h_i{}^0e_i{}^3]\\&+\frac{1}{(1-\nu)^2}\frac{1}{16}[5(1-(\mathbf{h}^0\cdot\mathbf{e})^2)+\sum_i e_i{}^4(1-5(\mathbf{h}^0\cdot\mathbf{e})^2)\\&\qquad-4\sum_i h_i{}^{02}e_i{}^2+8\mathbf{h}^0\cdot\mathbf{e}\sum_i h_i{}^{02}e_i{}^2]\Big\},\end{aligned}\tag{61}$$

with

$$\alpha=\tfrac{1}{6}(1-\sum_m b_m{}^{04})\quad\text{and}\quad\beta=\tfrac{1}{6}(5\sum_m b_m{}^{04}-3).$$

According to this, $j(\mathbf{e})$ depends explicitly on the direction of $\mathbf{e}$ with respect to the cubic axes and not only on the Bragg angle θ_B. Therefore, I_{00} and $I_{\mathbf{hh}}$ are in general

different, which leads to the asymmetry (54) of μ^{DS} and the total absorption (Fig. 3). The only exceptions are symmetrical reflections of ⟨100⟩ or ⟨110⟩ type. Furthermore, the absorption changes when, for a constant Bragg angle, the wave vector $\mathbf{k}$ is rotated around the $\mathbf{h}$ direction, as indicated in Fig. 2.

Equation (61) greatly simplifies if we average over all $\mathbf{b}^0$ directions, which is equivalent to assuming that loops occupy all planes with equal probability. Since $\langle\sum_m b_m{}^{04}\rangle = \frac{3}{5}$, $j(\mathbf{e})$ depends then only on $(\mathbf{h}\cdot\mathbf{e})^2=\sin^2\theta_B$ and $I_{00}{}^{ss}$ and $I_{\mathbf{hh}}{}^{ss}$ are equal. Then we have for the absorption with $F=\pi R_0^2$, and $P=1$ for σ, and $\cos 2\theta_B$ for π,

$$\mu^{DS}=\frac{1}{V_c}\left(\frac{e^2}{mc^2}Pf_{\mathbf{h}}'\right)^2\left(\frac{h}{k}\right)^2 C_L\left(\frac{b\pi R_0^2}{V_c}\right)^2\ln\frac{1}{q_cR_0}\times\left(\frac{8\pi}{15}+\frac{\pi}{15}\frac{-1+6\nu+3\nu^2}{(1-\nu)^2}\cos^2\theta_B\right). \quad (62)$$

This is equivalent to Eq. (51) for the cluster model. Most important is that μ^{DS} is proportional to $C_LR_0^4$ or to $cR_0^2[c=C_L(b\pi R_0^2/V_c)]$. Therefore, μ^{DS} increases due to the clustering by a factor of $(R_0/b)^2$ much faster than μ^{PE} which increases by R_0/b (43). The wavelength and h dependence of (62) is the same as for the cluster model, and analogously, we have to replace the cutoff q_c by $(q_0^2+q_c^2)^{1/2}$, if the Bragg condition is not exactly fulfilled, so that μ^{DS} and μ^{total} behave as illustrated in Fig. 1. The essential difference between formula (62) for loops and (51) for clusters is only that, for loops, μ^{DS} does not vanish for $\theta_B\to 90°$, which is due to the asymmetrical displacement field of a loop compared with the symmetrical field of the cluster.

The effect of lattice anisotropy can be taken into account by the perturbation theory[16,17] for G_{ij}. But the results for $j(\mathbf{e})$ are even more lengthy than (61) and will not be given. The effects of the anisotropic loop distribution are even enhanced by the lattice anisotropy.

VIII. DISCUSSION

We have shown that anomalous transmission is very sensitive to defect clustering if these clusters have a strong displacement field. For example, if a given number of point defects form dislocation loops of radius R_0, the photoelectric absorption μ^{PE} increases by a factor R_0/b and is proportional to $h^{3/2}$ (43). A similar result is obtained for the cluster model [(28) and (34)]. The diffuse scattering absorption μ^{DS} is even more sensitive to clustering and increases in the loop model by a factor $(R_0/b)^2$ (62) or by n_{cl} (51). Moreover, the diffuse scattering absorption is quite dissimilar to the photoelectric absorption with respect to the wavelength dependence ($\sim\lambda^2$) and the reflection dependence ($\sim f_{\mathbf{h}}'^2h^2$) [(51) and (62)]. Further, μ^{DS} depends explicitly on the direction of $\mathbf{k}$ (Fig. 2) and is more or less constant over the whole Bragg reflection (Fig. 1), leading to shoulders of the normal absorption, if one approaches the Bragg reflection.

A detailed comparison of this theory with an experiment of Baldwin *et al.*,[10] who measured anomalous transmission through Cu crystals after neutron irradiation at 40°C, has been published elsewhere.[28] In these crystals, dislocation loops are found ranging from 25 to 250 Å in diameter. Using the information which electron microscopy gives about the loop concentration and size distribution, μ^{PE} and μ^{DS} are calculated according to (43) and (62). For low concentrations, the theory agrees very well with experiment. For higher concentrations, the calculated values are systematically too low, which may be due to the counting problems of the electron microscope at high doses where the black spots begin to overlap.

To illustrate the importance of defect clustering, we have plotted in Fig. 4, on a double logarithmic scale, the absorption when a constant number of point defects build up dislocation loops of varying radius R_0. $\mu(R_0)$ divided by the absorption of fictitious loops of radius 1 Å is, therefore, essentially the absorption per point defect as a function of the loop radius R_0. The photoelectric absorption increases linearly with R_0, giving a straight line with slope 1 in Fig. 4, whereas the diffuse scattering absorption increases proportionally to R_0^2, giving a line with slope 2. As an example, we have taken the absorption of Mo $K\alpha$ and Cu $K\alpha$ radiation in Cu. The photoelectric absorption is accidentally nearly the same for both wavelengths ($\mu_0{}^{\text{Mo }K\alpha}=438$; $\mu_0{}^{\text{Cu }K\alpha}=446$ cm^{-1}), whereas, according to (62), the diffuse scattering absorption is a factor of $(\lambda_{Cu}/\lambda_{Mo})^2=4.5$ larger for Cu $K\alpha$ than for Mo $K\alpha$. For small radii, only the photoelectric absorption is important, which corresponds to the case of randomly distributed point defects,[6] the effects of which have been recently measured by Koehler *et al.*[29] For radii of the order of 50 Å, both absorption effects are of the same order of magnitude, in agreement with the experiment of Baldwin *et al.*[10] (neutron irradiaton at 40°C). For larger radii, the diffuse scattering absorption dominates and μ^{PE} becomes more and more negligible. Our theory is valid for radii $R_0\ll d_{\text{extinction}}$, say $R_0\lesssim 10^3$ Å. Therefore, the dashed lines above $R_0\gtrsim 10^3$ Å have only a qualitative meaning. But it seems to be plausible that the absorption has a maximum in the range of $R_0\approx d_{\text{ext}}$, because, for even larger loops, the x-rays can adjust themselves to the local-lattice distortions. Moreover, from the arguments given in Ref. 7, it seems to be plausible that the wavelength dependence is reversed for such large clusters. Such a wavelength sensitivity has been measured by Patel and Batterman,[7] who observed impurity oxygen clustering in Si, and by Hulett *et al.*,[30] who ob-

[28] F. W. Young, T. O. Baldwin, and P. H. Dederichs, in *Vacancies and Interstitials in Melals, Jülich, Germany, 1968* (North-Holland Publishing Co., Amsterdam, 1970).
[29] J. S. Koehler and L. Edelheidt (private communication).
[30] L. D. Hulett *et al.*, J. Appl. Phys. **39**, 3945 (1968).

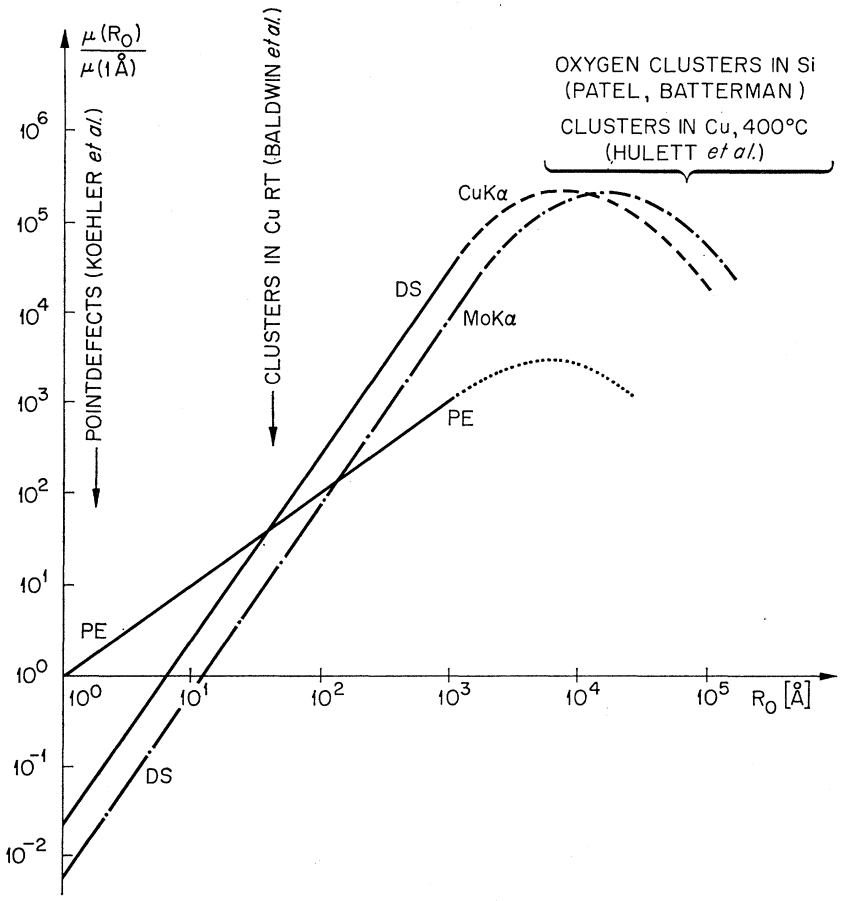

FIG. 4. Absorption "per point defect" as a function of the loop radius R_0.

served clusters in Cu, neutron irradiated at 400°C. In the latter experiment, the absorption per point defect was a factor 10^3 larger than in the 40°C irradiation experiment.[28]

From the scale of the ordinate, one sees that the absorption per point defect can increase by a factor $\sim 10^5$. Therefore, extremely small concentrations of point defects should be measurable if they are present in clusters of the "right" size. Hence, this theory may offer an explanation of the experiments of Efimov *et al.*,[8] who report the observation of the effect of $10^{14}/\text{cm}^3$ impurity hydrogen atoms on anomalous x-ray transmission in Ge.

Finally, we want to mention that the diffuse scattering from defect clusters in neutron-irradiated Cu crystals which gives rise to the extra absorption for the coherent wave has been directly observed recently in the Bragg geometry.[31] This diffuse scattering is extremely strong and concentrated within a few minutes of arc of the Bragg reflection. Moreover, the Bragg intensities are considerably reduced by the static Debye-Waller factor, which determines the photoelectric absorption in anomalous transmission. Therefore, anomalous x-ray transmission, as well as diffuse x-ray scattering, should prove to be useful to detect defect in clusters nearly perfect crystals.

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[31] T. O. Baldwin, P. H. Dederichs, and J. E. Thomas (unpublished).