Static electric field gradients and associated magnetic-resonance line-shape changes due to a random distribution of point defects*

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The distribution function for static electric field gradients arising from a random distribution of point defects in a crystal is studied. Results are obtained which include the magnitude as well as angular, concentration, and sample shape dependences for field gradients due to low concentrations of different types of point defects. The induced changes in frequency and time-domain spin-resonance line shapes are particularly simple if the only effect of the field gradients is to locally shift the spin energy levels. In this limit different types of defects cause characteristic line-shape changes which should be distinguishable.

I. INTRODUCTION

The effects on nuclear spins from electric field gradients arising from point defects have been treated by several investigators.[1,2] However, these treatments have not included an analysis of the angular dependence, concentration dependence, and magnitude of the effects arising from different types of point defects. In this paper we shall analyze the ways in which random distributions of different types of point defects change nuclear-spin line-shape functions. In some cases we predict that these changes depend upon the shape of the sample.

The task of calculating the effects of point defects on nuclear spins can be divided into two parts. The first part is finding the distribution of electric field gradients which arise from the point defects. This paper will be primarily concerned with obtaining this distribution function for small concentrations of defects. The second part of the problem is using the distribution function to find the changes in the nuclear-spin line-shape functions. For a small concentration of defects the changes are simple if the only effect of the electric field gradients is to shift the spin energy levels. In the present paper we shall work only in this limit.

In the remainder of this introduction we shall review some pertinent aspects of the theory of quadrupole splittings. In Sec. II we obtain a general expression for the distribution function of electric field gradients for various types of point defects at low concentrations. Section III contains a more detailed discussion for direct quadrupole fields arising from charged defects, induced quadrupole field gradients arising from charged defects, and electric field gradients arising from strains propagated by elastic forces.

Electric field gradients couple to the nuclear spins via the electric quadrupole Hamiltonian.[1] With an external magnetic field $\vec{H}_0$ acting on the spins, the first-order energy shift of the mth spin level arising from the electric field gradients is

$$E_m = [m^2 - \tfrac{1}{3} I(I+1)]\hbar\bar{\omega},$$

$$\bar{\omega} = A \sum_{i,j} V_{ij}(3\gamma_i\gamma_j - \delta_{ij}), \tag{1}$$

$$A = eQ' / 4I(2I - 1)\hbar.$$

In this equation the γ_i are the direction cosines of $\vec{H}_0$ with respect to the coordinate system of the crystal lattice, Q' is the nuclear quadrupole moment including the Sternheimer antishielding factor, and $V_{ij} = \partial^2 V/\partial x_i \partial x_j$, where V is the electric potential. It is assumed that the Zeeman splitting of the spin levels is large enough compared to the ΔE_m so that first-order perturbation theory is sufficient.

In this paper we shall assume that the only effect of the electric field gradients is to shift the spin energy levels and that the intrinsic spin decay rates are not changed. This is, of course, not true in general but it is a good approximation if the $|\Delta E_m/\hbar|$ are small compared to the intrinsic decay rates. Under this assumption the spectral shape function $G(\omega)$ can be written as

$$G(\omega) = \sum_m c_m g(\omega - a_m\bar{\omega}), \tag{2}$$

where $g(\omega)$ is the spectral shape function in the absence of the electric field gradients. The c_m's and a_m's are different for electromagnetic (NMR) transitions, nuclear-acoustic-resonance (NAR) $\Delta m = 1$ transitions, and NAR $\Delta m = 2$ transitions. Their values for different spin systems have been listed in various places[1-3] and will not be repeated here.

If a distribution of $\bar{\omega}$'s is given by $p(\bar{\omega})$, where $p(\bar{\omega})d\bar{\omega}$ is the probability that $\bar{\omega}$ lies between $\bar{\omega}$ and $\bar{\omega}+d\bar{\omega}$, then the observed spectral shape function in the frequency domain is the convolution

$$G(\omega) = \sum_m \int c_m d\bar{\omega}\, p(\bar{\omega}) g(\omega - a_m\bar{\omega}). \tag{3}$$

The corresponding spectral decay function in the time domain is

$$G(t) = \sum_m c_m g(t) Q(a_m t), \qquad (4)$$

where $G(t)$, $g(t)$, and $Q(t)$ are the Fourier transform of $G(\omega)$, $g(\omega)$, and $p(\overline{\omega})$, respectively.

Even when the spectral weight function does not take such a simple form, a calculation of $p(\overline{\omega})$ or $Q(t)$ is the first step in obtaining such functions. The formalism applies to electronic spins as well as nuclear spins with appropriate changes in numerical factors.

II. DISTRIBUTION FUNCTION

In this section we derive the distribution function for electric field gradients arising from a single type of point defect. Consider a lattice with N points and cN randomly distributed defects of the same kind where $\overline{\omega}(\vec{r}_\alpha)$ is the contribution to $\overline{\omega}$ [(see Eq. (1)] at the origin from a point defect at $\vec{r}_\alpha$. The value of $\overline{\omega}$ due to all of the defects is

$$\overline{\omega} = {\sum_\alpha}' \overline{\omega}(\vec{r}_\alpha), \qquad (5)$$

where the prime on the summation over lattice sites α means that only those lattice sites with defects are summed over. The probability that $\overline{\omega}$ lies between $\overline{\omega}$ and $\overline{\omega} + d\overline{\omega}$ is

$$p(\overline{\omega}) \propto \sum_k \delta\left(\overline{\omega} - {\sum_\alpha}' \overline{\omega}(\vec{r}_\alpha)\right), \qquad (6)$$

where the summation denoted by k is over all possible defect configurations. It is most convenient to normalize $p(\overline{\omega})$ later.

By expressing the δ function as an integral we obtain

$$p(\overline{\omega}) \propto \sum_k \int_{-\infty}^{\infty} dt\, e^{i\overline{\omega} t} \exp\left(-i \sum_\alpha [\overline{\omega}(r_\alpha)t]^{\delta_\alpha(k)}\right), \quad (7)$$

where $\delta_\alpha(k)$ equals 1 if the site α is occupied in the configuration k and is zero otherwise. Now note that

$$\sum_k \exp\left(-i \sum_\alpha [\overline{\omega}(\vec{r}_\alpha)t]^{\delta_\alpha(k)}\right) = \sum_k \prod_\alpha \exp^{-i\overline{\omega}(\vec{r}_\alpha)t\delta_\alpha(k)}.$$

$$(8)$$

However, because the distribution is random, the probability for occupation of a given site is independent of the occupation of all other sites and thus the expression in Eq. (8) is proportional to

$$\prod_\alpha [(1 - c) + c e^{-i\overline{\omega}(r_\alpha)t}]. \qquad (9)$$

By expressing the product as the exponential of a sum of logarithms we obtain

$$p(\overline{\omega}) = \int_{-\infty}^{\infty} dt\, Q(t) e^{i\overline{\omega} t},$$

$$Q(t) = \exp\left(\sum_\alpha \ln[1 - c(1 - e^{-i\overline{\omega}(\vec{r}_\alpha)t})]\right), \qquad (10)$$

which has the proper normalization.

For concentrations c much less than 1, this reduces to the more familiar expression[1,2]

$$Q(t) = \exp\left(\sum_\alpha c(e^{-i\overline{\omega}(\vec{r}_\alpha)t} - 1)\right). \qquad (11)$$

We note that if only a few nearest neighbors are included in the lattice summation, then $Q(t)$ equals 1 plus terms of order c. Thus for very small concentrations any short-ranged effects from the defects are insignificantly small and $Q(t)$ is dominated by the long-range part of $\overline{\omega}(\vec{r})$. This fact is partially obscured by the fact that all of the finite moments of $p(\overline{\omega})$ are dominated by the first few neighbors. Thus, except for insignificantly short times, the finite moments of $p(\omega)$ give a very poor picture of $Q(t)$. Since effects of any reasonable magnitude come from the far neighbors, the lattice is well approximated by a continuum. To an excellent approximation for $c \ll 1$, Eq. (11) can be written as

$$Q(t) = e^{-cI(t)}, \qquad (12a)$$

$$I(t) = \rho \int d^3 r (1 - e^{-i\overline{\omega}(\vec{r})t}), \qquad (12b)$$

where ρ is the density of lattice points per unit volume for the impurity under consideration and the integral is over the volume of the crystal.

If it is desired, the effects of the first few neighbors can be included by adding the appropriate term to $I(t)$. They will have the effect of producing an oscillatory noiselike contribution of order c to $Q(t)$. Similarly, an impurity whose $\overline{\omega}(\vec{r})$ cuts off exponentially will have very little effect on $p(\overline{\omega})$. These effects are rather obvious because if only a few spins are perturbed, the signal cannot change much.

In order to proceed further it is necessary to assume a functional form for $\overline{\omega}(\vec{r})$. In this section we consider only the form

$$\omega(r) = g(\Omega)/r^n, \qquad (13)$$

where $g(\Omega)$ is a real function of only the solid angle Ω and n is a positive number. In general $I(t)$ will have a real part $I_1(t)$ and an imaginary part $I_2(t)$. By expressing the volume integration as an angular integration times a radial intergration and making the substitution $x = r^{-n}$, one obtains

$$I_1(t) = \rho \int d\Omega \int_0^\infty \frac{dx}{nx^{1+\alpha}} [1 - \cos(gtx)], \qquad (14)$$

where $\alpha = 3/n$. This expression is convergent if $n > \frac{3}{2}$, which we now assume to be the case. The x integration is in standard integral tables and yields

$$I_1(t) = [\pi\rho |t|^\alpha / 2n\Gamma(1 + \alpha) \sin(\tfrac{1}{2}\pi\alpha)] \int d\Omega\, |g(\Omega)|^\alpha,$$

$$\alpha = 3/n, \quad n > \tfrac{3}{2}. \qquad (15)$$

The imaginary part of $I(t)$, which is ignored in most treatments, must be treated differently for $n > 3$, $n = 3$, and $n < 3$. For $n > 3$ there is no convergence problem and by making the substitution $x = r^{-n}$ one obtains

$$I_2(t) = \rho \int d\Omega \int_0^\infty \frac{dx}{nx^{1+\alpha}} \sin(gtx). \quad (16)$$

For $\alpha < 1$ ($n > 3$) this is a standard integral and yields

$$I_2(t) = [\rho\Gamma(1 - \alpha)\sin(\tfrac{1}{2}\alpha\pi)/3]$$

$$\times \int d\Omega \, |g(\Omega)t|^\alpha \epsilon(g(\Omega)t) ,$$

$$\alpha = 3/n, \quad n > 3, \quad (17)$$

where $\epsilon(x)$ is $+1$ if $x > 0$ and -1 if $x < 0$.

Even if $\int d\Omega g(\Omega)$ is zero, $I_2(t)$ is not necessarily zero. In fact if $g(\Omega)$ is proportional to any zonal harmonic except ones with $l = 1$, $I_2(t)$ will be nonzero. However, since $I_2(t)$ is an odd function of t, $p(\overline{\omega})$ is real. In addition $G(t)$ given by Eq. (4) is real since for every set (a_m, c_m) there is another set $(-a_m, c_m)$.

The case of $n = 3$ ($\alpha = 1$) must be treated more carefully. We shall assume that the angular average of $g(\Omega)$ is zero,

$$\int d\Omega \, g(\Omega) = 0. \quad (18)$$

In addition we exclude a small sphere of radius r_0 about the origin in Eq. (12b). This is appropriate because there are no neighbors at an arbitrarily close distance to the origin. Eventually we let $r_0 \to 0$ and the results are independent of r_0 in this limit. It is easiest to express Eq. (12b) as

$$I_2(t) = \rho \int_0 d^3r [\sin(gt/r^3) - (gt/r^3)]$$

$$+ \rho \int_0 d^3r (gt/r^3), \quad (19)$$

where the subscripts zero on a volume integration implies the limiting procedure described above. The first integral in Eq. (19) is convergent for large r and can easily be shown to be zero using Eq. (18). Thus we obtain

$$I_2(t) = \rho \int_0 d^3r \, [g(\Omega)t/r^3]. \quad (20)$$

Superficially $I_2(t)$ appears to vanish because of Eq. (18). However, because of the long-ranged $1/r^3$ factor in the integrand, it does not vanish. Evaluating Eq. (20) is very much like evaluating the demagnetization field in a ferromagnet.[4] An exact determination is possible only if the sample shape is an ellipsoid and $g(\Omega)$ is proportional to terms $(3\Omega_i\Omega_j - \delta_{ij})$, where Ω_i are direction cosines associated with the solid angle Ω. Even in this case $I_2(t)$ depends on the shape of the sample. For a

sample of arbitrary shape $I_2(t)$ depends on the position of the origin in the sample.

For $n < 3$ the situation is even more bizarre in that $I_2(t)$ depends not only on the sample shape but is actually proportional to the dimensions of the sample. We believe that such a situation is impossible to obtain in a charge neutral system. This point is further discussed in Sec. III.

Finally, in this section, we wish to point out that a field gradient that goes as $1/r^n$ will produce a $Q(t)$ that dies off exponentially as e^{-ct^α} and a $p(\overline{\omega})$ whose width is proportional to c^α, where $\alpha = 3/n$. In addition, the equations in this section are not necessarily restricted to substitutional or vacancy point defects. An interstitial impurity of a clump of impurities will often produce $\overline{\omega}(\vec{r})$'s that are of the form of Eq. (13) at large distances.

III. SPECIAL CASES

In this section we shall examine in detail some specific types of point defects.

A. Simple charged impurities

In a charge-neutral system with only one type of charged defect, the impurity charge must be screened in some fashion. In some cases the screening may be accomplished by conduction electrons and in other cases the screening may obtain via localized electrons. If the charge of the defect is screened out exponentially in a few atomic spacings, there will be very little effect on the quadrupole field distribution function. However, lattice distortions associated with the defect may propagate out as discussed in Sec. III C.

In many conductors an impurity of charge Ze is screened by conduction electrons so that the electric potential at large distances is given by[5]

$$V(\vec{r}) \sim Ze\xi \cos(2k_0r)/4k_f^2 r^3, \quad (21)$$

where k_0 is the Fermi wave vector of the conduction electrons and ξ is a dimensionless parameter which depends on the screening model used. At large distances this leads to electric field gradients[6]

$$V_{ij}(\vec{r}) = - Ze\xi\Omega_i\Omega_j \cos(2k_0r)/r^3, \quad (22)$$

where the Ω_i are the direction cosines of $\vec{r}$ with respect to the crystal axes. Because of the oscillatory behavior in r, this is not of the form of the field gradient considered in Sec. II. However, using Eqs. (1), (12b), and (22) we obtain

$$I(t) = \rho \int d\Omega \int_0^\infty r^2 dr \{1 - \exp[ig(\Omega)\cos(2k_0r)t/r^3]\},$$

$$(23)$$

$$g(\Omega) = \sum_{i,j} Ze\xi A(3\gamma_i\gamma_j - \delta_{ij})\Omega_i\Omega_j.$$

We cannot perform the radial integration in Eq. (23) in closed form. However, we can obtain an excellent approximation to it. The dominant con-

tributions to the radial integration comes from values of r near $r_0 = |gt|^{1/3}$. If $k_d r_0 \gg 1$, the r integration takes the integrand through many oscillation of $\cos(2k_0 r)$ while the rest of the integrand is slowly varying. This is the same as replacing $\cos(2k_0 r)$ by $\cos\phi$ and integrating ϕ from 0 to 2π, which yields

$$I(t) = \rho \int d\Omega \int_0^\infty r^2 \, dr [1 - J_0(g(\Omega)t/r^3)], \qquad (24)$$

where J_0 is the zeroth-order Bessel function. The r integration is now straightforward with the substitution $r = x^{-1/3}$ and yields

$$I(t) = \rho \int d\Omega \, \tfrac{1}{3} |g(\Omega)t|. \qquad (25)$$

However, from Eq. (12), $Q(t)$ is only significantly altered if $Ic \gtrsim \tfrac{1}{10}$ or $\rho|gt|c \gtrsim \tfrac{1}{10}$. Thus the condition that $k_0 r_0 \gg 1$ becomes $k_0 \rho^{-1/3} \gg c^{1/3}$. Since $k_0 \rho^{-1/3} \sim 1$ for most metals, this condition is easily satisfied for metals at low concentrations of charged impurities.

The remaining angular integration in Eq. (25) is proportional to

$$\int d\Omega \, |3(\vec{\gamma} \cdot \vec{\Omega})^2 - 1|, \qquad (26)$$

which is easily performed by choosing $\vec{\gamma}$ to define the polar axis and yields $16\pi/3^{3/2}$. Thus we obtain

$$I(t) = |16\pi Ze\xi\rho A/3^{5/2}| \, |t|. \qquad (27)$$

There is a set of conditions under which it is appropriate to use a potential of the form

$$V(r) = Ze/r. \qquad (28)$$

The conditions are that the charge-screening radius be much greater than $(\rho c)^{-1/3}$ and that the electric potential takes the form of Eq. (28) within that screening radius. In order to see this we note that the dominant part of the radial integration in Eq. (12) comes from values of r near $r_0 = |gt|^{1/3}$. In addition, since $I(t) \sim e^{-|\rho c g t|}$, it is only necessary that our approximation be good for times up to about $t \sim |10/\rho c g|$, or for values of $r_0 \sim |10/\rho c|^{1/3}$. Thus if the screening radius, which may depend on c itself, is much greater than $(\rho c)^{-1/3}$, Eq. (28) is a good approximation. However, since the interaction is ultimately screened, $I_2(t)$ as given by Eq. (20) vanishes.

Under the above conditions, we obtain

$$g(\Omega) = \sum_{i,j} AZe(3\gamma_i\gamma_j - \delta_{ij})(3\Omega_i\Omega_j - \delta_{ij}), \qquad (29)$$

where Z can take effective charges and dielectric constants into account. From Eq. (15) with $n = 3$,

$$I_1(t) = (\pi\rho|t|/6)\int d\Omega \, |g(\Omega)|. \qquad (30)$$

The remaining angular integration is identical to the one used in obtaining Eq. (27) and we obtain

$$I(t) = |8\pi^2 Ze\rho A/3^{3/2}| \, |t|. \qquad (31)$$

Both cases considered in this subsection generate spherically symmetric field gradients and thus $I(t)$ has no angular dependence.

B. Induced-field gradients

In some substances that have spin sites which lack inversion symmetry, electric fields can induce electric field gradients which are considerably larger than any direct effect.[7,8] The general connection between the electric field gradient and the electric field is written as

$$V_{ij} = \sum_k R_{ijk} E_k. \qquad (32)$$

For a zinc-blende structure, Eq. (32) can be written as[8]

$$V_{ij} = -R \sum_k \delta_{ijk} E_k. \qquad (33)$$

where δ_{ijk} is 1 if all three indices are different and is zero otherwise.

In this subsection we consider only electric fields arising from charged defects. We assume that the electric potential is $V(r) = Ze/r$ which is screened at long distances. Thus $\overline{\omega}(r)$ is of the form in Eq. (13) with $n = 2$ and

$$g(\Omega) = \sum_{i,j} AZe(3\gamma_i\gamma_j - \delta_{ij})R\delta_{ijk}\Omega_k. \qquad (34)$$

By arguments similar to those in Sec. III A, this is appropriate if the screening radius is much greater than $(\rho c)^{-1/3}$. In addition, $I_2(t)$ vanishes in this limit since Eq. (18) obtains. From Eq. (15) with $n = 2$ we obtain

$$I(t) = [(2\pi)^{1/2}\rho|t|^{3/2}/3]\int |g(\Omega)|^{3/2}d\Omega. \qquad (35)$$

The angular integration is proportional to

$$\int d\Omega \, |\vec{a} \cdot \vec{\Omega}|^{3/2}, \quad a_i = \sum_{j,k} \gamma_j\gamma_k\delta_{ijk}. \qquad (36)$$

The integral is easily performed by choosing $\vec{a}$ as the polar axis and yields $8\pi|a|^{3/2}/5$. Thus we obtain

$$I(t) = |(2\pi)^{3/2}AZeR\rho/15| \, |t|^{3/2}f(\vec{\gamma}),$$

$$f(\vec{\gamma}) = \left|\sum_{i \neq j} \gamma_i\gamma_j\right|^{3/2}. \qquad (37)$$

This has a characteristic angular dependence $f(\vec{\gamma})$ and is proportional to $|t|^{3/2}$. If we has used screening by conduction electrons, $I(t)$ would have been proportional to

$$\left| t \sum_{i \neq j} \gamma_i \gamma_j \right|. \tag{38}$$

C. Elastic strains

In this subsection we consider the effects of point defects which propagate strains and thus interact with the nuclear spins via the s tensor.[9] For a crystal of arbitrary symmetry the relationship between the field gradients and the strains is quite complicated and we shall only consider the case where cubic symmetry obtains

For a crystal with cubic symmetry,[9,10]

$$V_{ij} = [\tfrac{3}{2}(S_{11} - S_{44})\delta_{ij} + S_{44}]e_{ij} - \tfrac{1}{2}S_{11}\sum_k e_{kk}\delta_{ij},$$

$$e_{ij} = \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_j}\right)(1 - \tfrac{1}{2}\delta_{ij}), \tag{39}$$

where $\vec{u}(\vec{r})$ is the lattice displacement at the position $\vec{r}$. At large distances from an impurity site at the origin,[11] $u \sim 1/r^2$ and thus the strains go as $1/r^3$ and can be expressed as

$$e_{ij} = f_{ij}(\vec{\Omega})/r^3. \tag{40}$$

Thus $\overline{\omega}(\vec{r})$ is of the form given by Eq. (13) with $n = 3$ and

$$I_1(t) = (\pi \rho |t|/6) \int_0 |g(\Omega)| \, d(\Omega), \tag{41a}$$

$$I_2(t) = \rho \int_0 d^3 r [g(\Omega) t / r^3], \tag{41b}$$

$$g(\Omega) = 3AS_{44}\sum_{i \neq j}\gamma_i \gamma_j f_{ij} + \tfrac{3}{2}AS_{11}\sum_i (3\gamma_i^2 - 1)f_{ii}. \tag{41c}$$

Although a specific model is needed for the f_{ij}'s in order to obtain detailed results, there are some special directions for $\vec{H}_0$ which are particularly illuminating. For example, if $\vec{H}_0$ points along a cube edge, say in the z direction, then

$$g(\Omega) = \tfrac{3}{2}AS_{11}\left(3f_{zz} - \sum_i f_{ii}\right). \tag{42a}$$

In this case only S_{11} and compressional strains contribute to $I(t)$. Similarly, if $\vec{H}_0$ points along the [111] direction, then

$$g(\Omega) = AS_{44}\sum_{i \neq j} f_{ij} \tag{42b}$$

and only S_{44} and shear strains contribute to $I(t)$. These directions can be used to obtain the relative magnitude of shear and compressional strains. Further, if $|S_{11}f_{ii}|$ is much larger than $|S_{44}f_{ij}|$ with $i \neq j$, and $\vec{H}_0$ is in the plane defined by [001] and [110], then

$$g(\Omega) = \tfrac{3}{4}AS_{11}(3\cos^2\theta - 1)\left(3f_{zz} - \sum_i f_{ii}\right), \tag{42c}$$

where θ is the angle between $\vec{H}_0$ and [001]. In this case the angular dependence is quite simple.

In an isotropic elastic medium of infinite extent with isotropic defects,[11] $\vec{u} = B\vec{r}/r^3$, where B is a constant and thus

$$f_{ij} = B(3\Omega_i \Omega_j - \delta_{ij})(1 + \delta_{ij}).$$

A somewhat more general model that is also easy to work with is one where

$$f_{ij} = [f_c \delta_{ij} + 2f_s(1 - \delta_{ij})](3\Omega_i \Omega_j - \delta_{ij}). \tag{43}$$

In this model f_c and f_s refer to the compressional and shear part of the strains, respectively. If the medium is isotropic, then $f_s = f_c$. With this model, $g(\Omega)$ is

$$g(\Omega) = 18AS_{44}f_s \sum_{i \neq j} \Omega_i \Omega_j \gamma_i \gamma_j + \tfrac{3}{2}AS_{11}f_c \sum_i (3\gamma_i^2 - 1)$$

$$\times (3\Omega_i^2 - 1). \tag{44}$$

With the above model one can evaluate $I_1(t)$ numerically and $I_2(t)$ can be evaluated by the methods used for evaluating the demagnetization field in a ferromagnet. If $\vec{u}(\vec{r})$ really falls off as $1/r^3$ throughout a finite sample, then $I_2(t)$ depends on the sample shape. With the model described by Eq. (44), $I_2(t)$ can be evaluated exactly if the sample shape is an ellipsoid. If the sample shape is not an ellipsoid, then $I_2(t)$ for a given spin will depend on the position of that spin in the sample.

The shape and possible position dependence of $I_2(t)$ suggests quite a sensitive test of whether $u(\vec{r}) \sim 1/r^2$. Suppose, for example, that I_1 and I_2 can be evaluated and we obtain

$$I_1(t) = \overline{\omega}_1 |t|, \quad I_2(t) = \overline{\omega}_2 t, \tag{45}$$

where $\overline{\omega}_1$ is positive. Then

$$Q(t) = e^{-c\overline{\omega}_1|t| - ic\overline{\omega}_2 t} \tag{46a}$$

$$p(\overline{\omega}) = (c\overline{\omega}_1/\pi)/[(\overline{\omega} - c\overline{\omega}_2)^2 + (c\overline{\omega}_1)^2]. \tag{46b}$$

Equation (46b) describes a Lorentizian function with a width $c\overline{\omega}_1$ and centered at $c\overline{\omega}_2$. Previously it has been assumed that a Lorentizian line shape centered about some finite frequency is caused by some net strain in the crystal. This mechanism can be distinguished from the one which we are suggesting. If there is a net strain in some direction, then the line shape should be symmetirc about that direction. The symmetry in our model depends upon the shape of the crystal.

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