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Diffuse X-Ray Scattering from Crystalline Systems with Ellipsoidal Quantum Dots

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Abstract—A theory of diffuse X-ray scattering from a semiconductor system with ellipsoidal quantum dots (QDs) has been developed. The elastic strains outside a QD are calculated using the method of multipole expansions. An expression for the lattice displacement field is presented accurate to within the quadrupole term of expansion. Using the proposed approach, an analytical solution for the diffuse scattering from a crystalline medium with ellipsoidal inclusions is obtained. Reciprocal-space maps of the scattering intensity distribution are obtained by numerical calculations for QDs with various ratios of the height to lateral radius.

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High-resolution X-ray duffractometry (XRD) offers an effective approach to the investigation of various systems with self-organized quantum dots [1–4]. In recent years, much attention was devoted to determining the distributions of elastic strains around QDs (see, e.g., review [5] and references therein). These strains are usually calculated by numerical methods using either Green's function formalism or finite element method. This problem is difficult for an analysis and rarely yields analytical solutions. At the same time, the investigation of systems with QDs by high-resolution XRD techniques also requires one to construct models with allowance for the distribution of elastic strains. This task is even more complicated, since the it stipulates calculations of both the elastic strain fields and the diffuse X-ray scattering from the crystal lattice distortions caused by these strains. Evidently, the complexity of these problems accounts for the still small number of investigations in this direction [6–8].

As is known, a rather complete theory of diffuse X-ray scattering has only been developed for a model of crystal with spherically symmetric inclusions. However, this shape of inclusion is rather rare among self self-organized QDs. As a rule, the QD sizes in the lateral direction significantly exceed the vertical measurements. Previously developed models [6–8] considered QDs of cylindrical shape with allowance for the elastic strain and stress relaxation on the free surface. In these investigations, model solutions for calculating diffuse scattering were presented in the form of a product of the Fourier transform of the lattice displacement field of a point defect and the QD shape function. This approach is not quite correct, since the spatial variations of strains caused by spherical and cylindrical inclusions are different [9]. On the other hand, this difference is not as significant for defects of an ellipsoidal shape. Recently, ellipsoidal InAs quantum dots were observed by scanning tunneling microscopy [10].

Nenashev and Dvurechenskii [11] suggested to calculate the QD-induced strains using an analogy between the problems of electrostatics and the theory of elasticity. According to this approach, the vector of elastic displacements at a point **r** in the crystalline medium, which is related to an inclusion with an arbitrary shape can be written as follows:

$$\delta\mathbf{u}(\mathbf{r}) = \Lambda\int_V \frac{\mathbf{r}-\mathbf{r}'}{|\mathbf{r}-\mathbf{r}'|^3}d\mathbf{r}, \tag{1}$$

where $\Lambda = \varepsilon_0(1+\nu)/[4\pi(1-\nu)]$; $\varepsilon_0 = (a_{\text{inclusion}} - a_{\text{matrix}})/a_{\text{matrix}}$ is the relative lattice mismatch between the inclusion ($a_{\text{inclusion}}$) and matrix (a_{matrix}), ν is the Poisson ratio, and V is the inclusion volume. Let us define the potential of a homogeneous inclusion as follows [11]:

$$\varphi(\mathbf{r}) = \Lambda\int_V \frac{d\mathbf{r}'}{|\mathbf{r}-\mathbf{r}'|}, \tag{2}$$

which can be calculated using the method of multipole expansion as

$$\begin{aligned}|\mathbf{r}-\mathbf{r}'|^{-1} &= (r^2 - 2rr'\cos(\gamma) + r'^2)^{-1/2}\\ &= \sum_{n=0}^{\infty}\frac{r'^n}{r^{n+1}}P_n(\cos(\gamma)),\end{aligned} \tag{3}$$

where $P_n(\cos(\gamma))$ are the Legendre polynomials and γ is the angle between vectors $\mathbf{r}$ and $\mathbf{r}'$. In terms of this expansion, the potential (2) can be expressed by the following series:

$$\varphi(\mathbf{r}) = \varphi_0(\mathbf{r}) + \varphi_1(\mathbf{r}) + \varphi_2(\mathbf{r})\ldots\varphi_n(\mathbf{r})\ldots, \qquad (4)$$

where

$$\varphi_n(\mathbf{r}) = \Lambda\frac{P_n(\cos(\gamma))}{r^{n+1}}\Omega_n,$$

$$\Omega_n = \int\limits_V r'^n d\mathbf{r}'.$$

Here, $\varphi_0(\mathbf{r})$ describes the potential of a point or spherically symmetric field; $\varphi_1(\mathbf{r})$ and $\varphi_2(\mathbf{r})$ are the dipole and quadrupole terms, respectively; etc.

According to expressions (1) and (2), the lattice displacement field due to a QD is determined by the potential gradient $\delta\mathbf{u}(\mathbf{r}) = -\nabla\varphi(\mathbf{r})$ and, hence, can also be expressed by a series as

$$\delta\mathbf{u}(\mathbf{r}) = \delta\mathbf{u}_0(\mathbf{r}) + \delta\mathbf{u}_1(\mathbf{r}) + \delta\mathbf{u}_2(\mathbf{r}) + \ldots\delta\mathbf{u}_n(\mathbf{r})\ldots, \qquad (5)$$

where $\delta\mathbf{u}_n(\mathbf{r}) = \Lambda(n+1)\dfrac{\mathbf{r}P_n(\cos\gamma)}{r^{n+3}}\Omega_n$.

For a point (or spherically symmetric) source of elastic stresses, a solution of type (1) is well known as the lattice displacement field of Coulomb defects [12], which is determined by the main term of expansion (5) as shown below:

$$\delta\mathbf{u}(\mathbf{r}) = \delta\mathbf{u}_0(\mathbf{r}) = \begin{cases} \text{random}, & |\mathbf{r}| \le R, \\ \Lambda\dfrac{4\pi R^3}{3}\dfrac{\mathbf{r}}{r^3}, & |r| > R. \end{cases} \qquad (6)$$

where R is the radius of the spherical inclusion.

Now let us consider a model of an ellipsoidal QD (Fig. 1) with the vertical ellipsoid axis l_z and the horizontal radius R ($2R$ being the lateral ellipsoid axis). According to this model, the elastic displacements can be expressed (to within the quadrupole term of the multipole expansion) as follows:

$$\delta\mathbf{u}(\mathbf{r}) \approx \begin{cases} \text{random}, & |z| \le |l_z/2|, \quad \sqrt{x^2+y^2} \le R, \\ A\dfrac{\mathbf{r}}{r^3} + B\dfrac{\mathbf{r}(3\cos^2(\theta)-1)}{r^5}, & |z| > |l_z/2|, \quad \sqrt{x^2+y^2} > R, \end{cases} \qquad (7)$$

where $A = \Lambda V_{\text{ell}}$ is the strength of the ellipsoidal QD, $V_{\text{ell}} = (2\pi/3)l_zR^2$ is the volume of the ellipsoid, and $B = -3A[R^2 - (l_z/2)^2]/25$. Unlike formula (6), the elastic displacements of ellipsoidal QDs depend on the angle θ between the z axis and r direction (Fig. 1). In the case of $l_z = 2R$, the ellipsoid transforms into a sphere, coefficient B vanishes, and solution (7) coincides with formula (6).

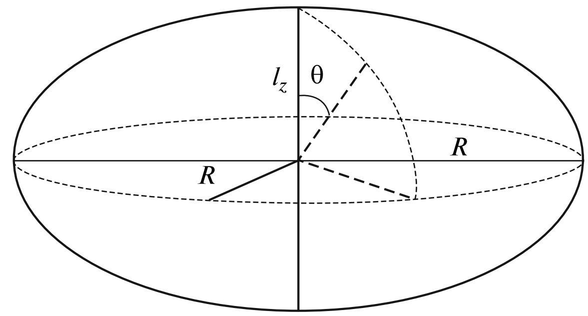

Fig. 1. Model of an ellipsoidal QD (see text for explanations).

Let us write the intensity of diffuse X-ray scattering as

$$I_h^d(\mathbf{q}) = K_D|D(\mathbf{q})|^2, \qquad (8)$$

where K_D is a constant coefficient [13] and $D(q)$ is the diffuse scattering amplitude that can be written as follows:

$$D(\mathbf{q}) = D_{\text{SW}}(\mathbf{q}) + D_{\text{H}}(\mathbf{q}) + D_{\text{Q}}(\mathbf{q}). \qquad (9)$$

Here, $D_{\text{SW}}(\mathbf{q})$ is the amplitude of scattering for ellipsoidal inclusions with neglect of strains outside the QD (i.e., the Stokes–Wilson scattering amplitude), $D_{\text{H}}(\mathbf{q})$ is the Huang scattering amplitude, and $D_{\text{Q}}(\mathbf{q})$ is the amplitude correction due to the allowance for the quadrupole term in the strain field.

The Stokes–Wilson scattering amplitude for a model ellipsoidal inclusion was calculated in a cylindrical coordinate system as

$$D_{\text{SW}}(\mathbf{q}) = 2\pi\int\limits_{-l_z/2}^{l_z/2}\frac{R_z}{q_0}I_1(q_0R_z)\exp(iq_zz)dz, \qquad (10)$$

where $J_1(q_0R_z)$ is the first-order Bessel function, $q_0 = \sqrt{q_x^2+q_y^2}$, and $R_z = R\sqrt{1-z^2/(l_z/2)^2}$. The second term of sum (9), which describes the Huang scattering, was calculated in the generalized spherical coordinate system. The final expression for the amplitude of this diffuse scattering is as follows

$$D_{\text{H}}(\mathbf{q}) = \frac{2\pi A\mathbf{hq}}{q^2}\Phi_1(q, R, l_z), \qquad (11)$$

where $\Phi_1(q, R, l_z)$ is a function that depends on $q = \sqrt{q_x^2+q_y^2+q_z^2}$ and the parameters R and l_z of the ellipsoidal QD.

$$\Phi_1(q, R, l_z) = \int\limits_{-1}^{1}dx\exp(iq\rho(x, l_z)x),$$

and

$$\rho(x, l_z) = R\left(1 + \left[\frac{R^2}{(l_z/2)^2} - 1\right]x^2\right)^{-1/2}.$$

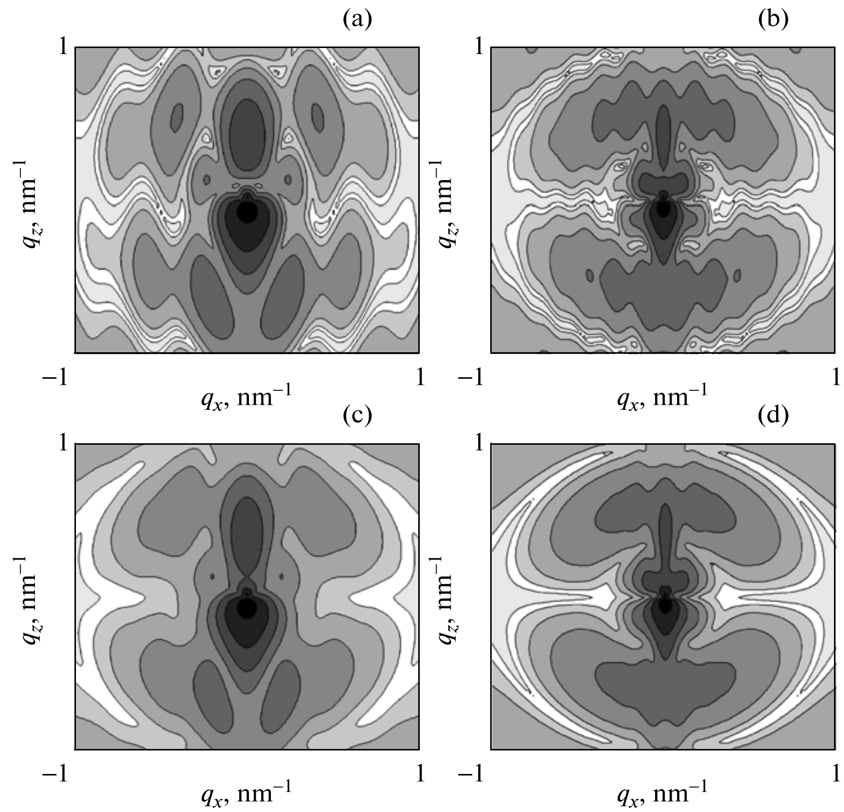

Fig. 2. RSMs of intensity of diffuse scattering from crystalline medium with randomly distributed ellipsoidal QDs with height of $l_z = 10$ nm, and lateral size (a, c) $R = 20$ nm and (b, d) 40 nm in the case (a, b) where all QDs have identical sizes and (c, d) QDs distributed with 30% variance of vertical and lateral size fluctuations.

The quadrupole correction for $R \le 4l_z$ is much smaller than the first two terms in formula (9) and can be written by analogy with relation (11) as follows:

$$D_Q(\mathbf{q}) = 6\pi A \frac{[R^2 - (l_z/2)^2]\mathbf{hq}}{25}\Phi_2(q, R, l_z), \qquad (12)$$

where

$$\Phi_2(q, R, l_z) = \int_{-1}^{1} dx(3x^2-1)x^2\left[\frac{e^{iqR(x)x}}{iqR(x)x} + E_1(-iqR(x)x)\right],$$

$$E_1(iR) = \int_R^{\infty} dz \frac{\exp(-iz)}{z}$$

is integral exponent.

Using the model solution expressed by Eqs. (8)–(12), we have numerically simulated the diffuse X-ray scattering intensity in the reciprocal space for a crystalline medium with randomly distributed ellipsoidal QDs. For the sake of simplicity, let us assume the medium to be infinite, ignore the stress relaxation on the free surface, and neglect any spatial correlation in the arrangement of QDs.

Figure 2a shows the results of calculations of the of the diffuse scattering intensity distribution near the reciprocal lattice node (reciprocal space maps, RSMs) for a crystalline medium with randomly distributed ellipsoidal QDs of the same height of 10 nm. Figure 2a presents the pattern of equal intensity lines (isolines) for a system with QDs of the same lateral size (radius) of $R = 20$ nm. In this and all other RSM maps, the ratio of intensities between the neighboring isolines is plotted on a logarithmic scale and amounts to 0.237. Since the lateral size for these QDs is four times the height, the patterns of diffuse scattering significantly differs from that obtained for a medium with spherically symmetric inclusions [13]. This is manifested by narrowing of the angular distribution and by its clearly pronounced oscillatory character. The further increase in the lateral size (to $R = 40$ nm) even stronger changes the shapes of isolines (Fig. 2b) as compared to those for a system with spherical inclusions.

In the course of epitaxial growth, it is impossible to ensure the formation of absolutely identical self-organized QDs and, hence, it is necessary to perform a procedure of averaging over the sizes of inclusions. The results of this procedure are presented in Figs. 2c and 2d.

It should be emphasized that, based on numerical estimations, it can be concluded that the pattern of diffuse X-ray scattering is determined primarily by the first two terms of Eq. (9). The contribution of the quadrupole term in the expansion of the lattice displacement field is extremely small in magnitude and resembles the angle distribution of the Huang scattering.

In conclusion, it should be noted that, although the elastic strain fields for ellipsoidal inclusions can be calculated using an analytical solution [14], the calculation of diffuse scattering based on this solution is not less difficult and requires no shorter computation time than, e.g., the numerical simulation procedure based on the finite element method.

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