# Influence of a glass matrix on acoustic phonons confined in microcrystals

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The influence of a glass matrix on the spheroidal and torsional vibrational modes of microcrystals embedded into the matrix is analyzed. The low-frequency Raman scattering data are used to show that the influence of the matrix on the frequencies of the acoustic phonons confined in the microcrystals is significant even if Lame's constants  $\lambda$  and  $\mu$  and mass densities of a microcrystal and a matrix are quite different from each other. It was found that surface vibrations arise as microcrystals are embedded in a matrix. These vibrations are caused by the restoring force which limits the free rotation of microcrystals. Most likely, just these vibrations take part in the low-frequency Raman scattering described in a number of papers.

#### I. INTRODUCTION

Many of the anomalous thermal and electrical properties of microcrystals are attributable to the influence of surface and size effects on the phonon spectrum of these particles. In semicondutor microcrystals, not only the electronic but also the lattice vibrational modes become discrete due to threedimensional confinement. As the size of microcrystals decreases, phonons with a larger wave vector are involved in the electron-phonon interaction. Hence, the electron-phonon interaction with acoustic phonon modes becomes more pronounced as compared to the scattering of electrons by optical phonons. It was found, for example, that electron-phonon interactions with acoustic phonons were found to be responsible for the decay rate of the excitonic polarization in semiconductor microcrystals.<sup>1</sup> It turned out that size-quantized acoustic phonon modes in microcrystals can be observed with the help of low-frequency Raman scattering in the range of the spectrum  $\sim 10 \text{ cm}^{-1.2}$  After that, similar lowfrequency Raman scattering spectra for various microcrystals, embedded in a glass matrix, were reported by other authors.<sup>3-7</sup> In all these papers, on the basis of the fact that Lame's constants  $\lambda$  and  $\mu$  and the mass density  $\rho$  of the microcrystals and a matrix are much different from each other, experimental data are interpreted under the assumption that the microcrystal surface is free.

In the present paper, using the theory of Tamura *et al.*,<sup>8</sup> we consider the influence of the matrix on the spheroidal and torsional vibrational modes of microcrystals, which brought about an interpretation of the low-frequency Raman spectra. We have studied the heterophase system, consisting of Ge microcrystals embedded in a GeO<sub>2</sub> glass matrix.<sup>4</sup> To gain a better understanding of the influence of the crystal-matrix boundary we have also analyzed some other chemical compositions of microcrystals and matrices with different relations between elastic constants and densities such as CdS in a GeO<sub>2</sub> matrix and Ag in a SiO<sub>2</sub> matrix which were obtained in Refs. 6,7. We have found that surface vibrations arise when microcrystals are embedded in a matrix. These vibrations are due to the restoring force which limits the free

rotation of microcrystals. Most likely, just these vibrations take part in the low-frequency Raman scattering described in a number of papers.<sup>4–7</sup> Also, it was found that the matrix had a different influence on the eigenfrequencies of the spheroidal and torsional vibrational modes. It is related to the fact that the torsional modes are purely transverse whereas the spheroidal modes are mixed modes of transverse and longitudinal nature. Here we show that one cannot disregard the influence of the matrix even if Lame's constants  $\lambda$  and  $\mu$  and the mass density  $\rho$  of microcrystals and a matrix are very different from each other.

#### **II. EXPERIMENT**

In our work<sup>4</sup> it was shown that experimentally, by choosing the proper technological conditions, germanium quantum dots, embedded in the solid transparent matrix, were obtained. We are able to produce transparent films of GeO<sub>2</sub> with Ge microcrystals embedded in them with given sizes from 5 nm to the greater using the reaction  $2\text{GeO}(\text{gas}) = \text{Ge} + \text{GeO}_2$ . The ratio between amorphous and crystalline phases of the germanium microparticles depends on the film growth temperature, as appears from the Raman scattering spectra. The structure of a heterofilm (the amount of the amorphous and crystalline phases of the germanium microparticles, the particles sizes, the transformation Ge to GeO<sub>2</sub>, and removal of the amorphous matrix GeO<sub>2</sub> from the film) could be modified by special treatments such as annealings, oxidation, and etching.

Raman measurements were carried out in a conventional 90° scattering geometry at room temperature. The spectra were excited by the 514.5- and 647.1-nm lines of Ar-ion and Kr-ion lasers and recorded by a Jobin Yvon U-1000 spectro-photometer equipped with a double monochromator and photon-counting system.

## **III. RESULTS AND DISCUSSION**

# A. Eigenfrequencies of torsional vibrational modes of microcrystals

In Ref. 9 it was shown that due to selection rules for Raman scattering on spherical particles, only spheroidal

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modes with even angular momentum quantum numbers l can be seen in the Raman spectrum. However, in practice the form of the microcrystals in a glass matrix may have deviations from a perfect spherical one. In this case, the selection rules can be relaxed, and not only spheroidal modes with even l, but torsional vibrational modes with odd l can be seen in the Raman spectra. According to Ref. 8, eigenfrequencies of the torsional vibrational modes of the microcrystals embedded in a glass matrix can be determined from the equation

$$D_m^t = 0, \tag{1}$$

where  $D_m^t$  is the determinant of the following matrix:

$$\left( \begin{array}{c} \frac{(l-1)}{\eta_{i}} j_{l}(\eta_{i}) - j_{l+1}(\eta_{i}) & j_{l}(\eta_{i}) \\ C_{1}C_{2} \sqrt{\frac{\pi}{2 \eta_{m}}} \left[ \frac{(l-1)}{\eta_{m}} K_{l+1/2}(\eta_{m}) - K_{l+3/2}(\eta_{m}) \right] & \sqrt{\frac{\pi}{2 \eta_{m}}} K_{l+1/2}(\eta_{m}) \end{array} \right)$$

where  $j_l(\eta)$  is the spherical Bessel function of the first kind,  $\eta_{n,l} = \omega_{n,l} d/2v_t$  is the dimensionless frequency,  $\omega_{n,l}$  is the angular frequency of (n,l) vibrational modes, l is the angular momentum quantum number, n is the number of the harmonic, and d is the diameter of a microcrystal. We obtain the angular frequencies  $\omega_{n,l}$  from the eigenvalues  $\eta_{n,l}$  and then arrive at the Raman-shift wave numbers we  $\nu_{n,l} = \omega_{n,l}/2\pi c$ , where c is the velocity of light.  $\sqrt{\pi/2\eta}K_{l+1/2}(\eta)$  is the modified spherical Bessel function of third kind,  $\eta_m = \eta_i v_{ti} / v_{tm}, \qquad C_1 = \sqrt{\mu_m / \mu_i},$ the  $C_2 = \sqrt{\rho_m}/\rho_i$ ,  $v_t$  is the transverse sound velocity,  $\mu$  is Lame's constant, and  $\rho$  is the mass density. The index m corresponds to the matrix and *i* to a microcrystal. Let us note that Ref. 8 reports erroneous terms  $d_{21}$  and  $d_{22}$  of the matrix  $D_m^t$ . In Fig. 1(a) the shift of vibrational eigenfrequencies



FIG. 1. (a) The shift of the eigenfrequencies of the torsional vibrational modes of the microcrystals for the surface mode (n=0) and the inner modes (n=1,2) as a function of the matrix stiffness, i.e., as a function of the ratio  $C_1/C_2$  at fixed  $C_2=0.77$ . The dashed lines correspond to the case of the free microcrystal surface. The discrete values of the frequencies for different angular momenta *l* are joined by the line which is a guide for an eye. (b) The radial distribution of the vibrational amplitudes *u* and the energy density  $u^2R^2$  of the surface vibrational mode of a microcrystal as a function of the ratio  $C_1/C_2$  at fixed  $C_2$ .

 $\eta_{nl}^T$  of a microcrystal is shown as a function of the ratio  $C_1/C_2$  at fixed  $C_2$ . In other words, it is the evolution of a shift when the matrix stiffness increases relative to that of the microcrystal. In Fig. 1(b) the radial distribution of the vibrational amplitudes u and the energy density  $u^2R^2$  of the surface vibrational mode are shown as a function of the ratio  $C_1/C_2$ . Figure 1(a) shows that the transverse surface mode (n=0) with the angular momentum l=1 is absent for microcrystals with a free surface (dashed lines). This is natural, because this mode corresponds to the rotation of the microcrystal as a whole. If a microcrystal is embedded in a matrix, then there appears a new solution for the surface mode with the angular momentum l=1. This mode arises because of the restoring force which limits the free rotation of the microcrystal.

Figure 1 shows that the surface vibrational mode of the microcrystals embedded in the matrix (i.e., the mode with n=0) does not disappear; its eigenfrequency increases and the maximum of the vibrational amplitude shifts inside the microcrystal with increasing of the matrix stiffness. The same happens with the inner vibrational modes  $(n \ge 1)$ , as it was pointed out earlier in Ref. 5. In Ref. 8 an erroneous conclusion was made that the influence of the matrix over the microcrystal leads to the opposite effect; i.e., the surface mode disappears and the inner vibrational modes soften. Hence, contrary to Refs. 6,7, the frequency of the Raman peak for the surface torsional mode with angular momentum l=1 for the microcrystals embedded the matrix does not equal  $v_1^T = 1.83 v_t / dc$  as would be in the case of the free surface. Indeed, the existence of the matrix results in the appearance of a new solution of (1) for the surface mode with l=1; its eigenfrequency increases, for example, from 0.2  $v_t/dc$  to 1.4  $v_t/dc$  when the matrix stiffness  $C_1/C_2$ increases 0.1-3.5 [Fig. 1(a)].

## B. Eigenfrequencies of spheroidal vibrational modes of microcrystals

Eigenfrequencies of the spheroidal vibrational modes of the microcrystals, embedded the matrix, can be determined from the equation

$$D_m^s = 0, (2)$$



FIG. 2. The distribution of the vibrational eigenfrequencies for the spheroidal and torsional vibrational modes of the microcrystals embedded in the glass matrices. The dashed lines correspond to the case of the free microcrystal surface for n = 0 and 1.

where  $D_m^s$  is the determinant of a (4×4) matrix which is given in the Appendix. In this case the number of variables and parameters is twice as large in comparison with the case of the torsional vibrations. Respectively, the influence of the matrix on the eigenfrequencies of the spheroidal vibrational modes of a microcrystal becomes more intricate. First of all, in this case, just as in the case of torsional modes, a new solution for the surface mode (n=0) with l=1 can arise [see Fig. 2(b)]. The frequency of this mode increases with increasing stiffness of the matrix in such a way that the mode with frequency  $v_1^{S} = 1.17 v_t/dc$ , which in the absence of the matrix was a surface mode with  $n = 0,^{6,7}$  now becomes an inner mode with n = 1. The other surface modes, with angular momenta  $l=0,2,3,\ldots$ , show a different behavior as compared to the torsional surface modes. They are softened when the stiffness of the matrix increases (see Fig. 2) and can disappear completely at some definite values of the elastic constants of microcrystals and matrices. Usually it is assumed that microcrystals can be considered as free when Lame's constants  $\lambda$ ,  $\mu$  and the mass density  $\rho$  of a microcrystal and a matrix are quite different from each other. We believe that according to our results, this assumption is incorrect. The torsional surface modes do not disappear even at close values of the parameters  $\lambda$ ,  $\mu$ , and  $\rho$  of a microcrystal and a matrix, but the spheroidal surface modes  $\nu_0^S$  (l=0) and  $\nu_2^{S}$  (l=2), which are active in Raman scattering, have a tendency to disappear at quite different values of the parameters. For example, in case of Pb microcrystals in porous glass<sup>8</sup> the mode  $\nu_0^S$  disappeared. It would exist if  $\mu_m \leq \mu_i$ ,  $(C_1 \leq 1)$ , but at  $\mu_m > \mu_i$ ,  $(C_1 > 1)$  the solution disappears, although all parameters except  $\mu$  for microcrystals and a matrix are drastically different. The mode  $\nu_2^S$  becomes softer after embedding Pb particles in a SiO<sub>2</sub> matrix, but it does not disappear completely as is mentioned in Ref. 8; it softened from  $0.85v_t/dc$  to  $0.6v_t/dc$ .

### C. Comparison of theory to previous experimental results

Let us consider now the interpretation of low-frequency Raman scattering on the CdS microcrystals embedded a GeO<sub>2</sub> matrix.<sup>6</sup> In this paper it was assumed that the surface of the microcrystals is free. The most distinct spectra were obtained on microcrystals with d = 7.5 nm. For parallel polarization the frequency of the maximum was  $v_{|| ||} = 14.5$ cm<sup>-1</sup>, while for the crossed polarization  $v_{||\perp} = 7.1$  cm<sup>-1</sup>. The authors have assumed that the totally polarized signal at 14.5 cm<sup>-1</sup> can arise only from the spheroidal inner mode with l=0, n=1 and that the signal from the surface mode with n=0 at 6 cm<sup>-1</sup> is masked by diffusion scattering. According to their estimation, the authors ascribed the signal at 7.1 cm<sup>-1</sup> to the surface mode with l=2. We have recalculated the eigenfrequencies of the microcrystals CdS in a GeO<sub>2</sub> matrix at the following values of the parameters:

$$\rho_{CdS} = 4.82 \text{ g/cm}^3$$
,  $v_l = 4.25 \times 10^5 \text{ cm/sec}$ ,  $v_t = 1.86 \times 10^5 \text{ cm/sec}$  (Ref. 6),  
 $\rho_{GeO_2} = 3.6 \text{ g/cm}^3$ ,  $v_l = 3.43 \times 10^5 \text{ cm/sec}$ ,  $v_t = 2.14 \times 10^5 \text{ cm/sec}$  (Ref. 10)

Coefficients in the determinants  $D_m^s$  and  $D_m^t$  were found to be  $C_1 = 0.99$ ,  $C_2 = 0.86$ ,  $C_3 = 2.28$ ,  $C_4 = 1.6$ ; the results of the estimation are shown in Fig. 2(a). As was mentioned above, the spheroidal surface modes with l=0 must disappear when the parameter  $C_1$  goes to 1. In the present case the mode  $v_0^S$  softened from  $0.78v_t/dc$  to  $0.35v_t/dc$  and became equal to 2.9 cm<sup>-1</sup> [Fig. 2(a)]. It means that the authors of Ref. 6 were not able to observe this mode. The next inner modes with l=0, as seen in Fig. 2(a), are situated at  $1.69v_t/dc$  and  $1.94v_t/dc$ , i.e., at 14 and 16 cm<sup>-1</sup>, respectively. They correspond to the experimental value 14.5 cm<sup>-1</sup>. However, one has to remember that the strength of

the light interaction with higher harmonics sharply decreases, because under the relaxation of the wave vector selection rules, which takes place in microcrystals, the spectrum of the wave vectors of the phonons becomes Lorentzian instead of a  $\delta$  function near the center of the Brillouin zone. Contrary to that, in Ref. 6 the intensities of a signal for the surface harmonic at 7.1 cm<sup>-1</sup> and for the inner harmonic at 14.5 cm<sup>-1</sup> are approximately equal. The second reason which leads to suppression of the inner harmonics in the Raman spectrum is the broad size distribution of microcrystals. It results in a large half-width of the experimental spectrum. For example, we have observed the inner harmonics on a single epitaxial film with a half-width of the signal equal to 5 cm<sup>-1</sup> (Ref. 11); in Ref. 6 the half-width of the signal is 10 cm<sup>-1</sup>.

Let us discuss now the depolarized signal at 7.1 cm<sup>-1</sup>. The spheroidal surface mode with l=2 was also softening from  $0.85v_t/dc$  to  $0.35v_t/dc$  and became equal to 2.9 cm<sup>-1</sup>, Fig. 2(a). The authors of Ref. 6 could not also observe this mode. Most probably, the mode used in Ref. 6 at 7.1 cm<sup>-1</sup> is the torsional surface mode with l=1, which occurred when the microcrystal was embedded in a matrix and which can be observed due to deviation of the microcrystal shape from the ideal spherical form. As one can see in Fig. 2(a), the frequency of the surface torsional mode is equal to

 $\nu_1^T = 1.18 \upsilon_t / dc = 9.7 \text{ cm}^{-1}$ . This is somewhat higher than 7.1 cm<sup>-1</sup>; however, we have to take into account the fact that the spectra are measured in the presence of a strong diffusional scattering which leads to a shift of the peak maximum in the direction of low frequencies. After subtracting the diffusional scattering from the experimental spectrum the frequency of the maximum must shift from 7.1 cm<sup>-1</sup> to the theoretical value 9.7 cm<sup>-1</sup>.

Let us consider now the case of Raman scattering on Ag microcrystals embedded in a SiO<sub>2</sub> matrix given in Ref. 7. It was also assumed in this paper that the microcrystal's surface is free. This assumption is based on the fact that the parameters  $\lambda$  and  $\rho$  for Ag are very different from those for SiO<sub>2</sub>. In Ref. 7 it was pointed out that for microcrystals with a size larger than 4.0 nm the agreement between the theory and the experiment is poor. In the experimental spectra for particles of size 4.1 nm the position of the maximum is equal to 14.2 cm<sup>-1</sup> and for particles of size 5.2 cm<sup>-1</sup> is equal to 12.1 cm<sup>-1</sup>. The authors assumed that these spectra are caused either by surface spheroidal modes with l=0 or by spheroidal modes with l=2, as they are partially depolarized.

We calculated eigenfrequencies of the vibrational modes of Ag microcrystals in a  $SiO_2$  matrix at the following values of parameters:

 $\rho_{Ag} = 10 \text{ g/cm}^3$ ,  $v_l = 3.65 \times 10^5 \text{ cm/sec}$ ,  $v_t = 1.66 \times 10^5 \text{ cm/sec}$  (Ref. 7),  $\rho_{SiO_2} = 2.2 \text{ g/cm}^3$ ,  $v_l = 5.95 \times 10^5 \text{ cm/sec}$ ,  $v_t = 3.76 \times 10^5 \text{ cm/sec}$  (Ref. 12).

Coefficients in the determinants  $D_m^s$  and  $D_m^t$  are found to be equal to  $C_1 = 1.06$ ,  $C_2 = 0.47$ ,  $C_3 = 2.2$ ,  $C_4 = 1.5$ . The results of the estimation are shown in Fig. 2(b). Here, as in the previous case, the parameter  $C_1$  is close to 1; it means that the surface spheroidal vibrations with l=0 must damp. Figure 2(b) shows that the frequency  $\nu_0^S$  softens from  $0.78v_t/dc$  to  $0.2v_t/dc$  and is equal to 2.7 cm<sup>-1</sup> for particles with d = 4.1 nm and 2.4 cm<sup>-1</sup> for particles with d = 5.2 nm. From these results one can conclude that these modes cannot be observed because they are masked by diffusional scattering. The spheroidal surface vibrations with l=2 also soften and have an eigenfrequency much smaller than the experimental value, Fig. 2(b). We come to the conclusion that the only possible suggestion is that these vibrations are torsional surface modes with l=1 which appear when the crystals are embedded in a matrix and are active in Raman scattering due to deviations of the microcrystal shape from a spherical one. As one can see from Fig. 2(b), for particles with d=4.1 nm the frequency of the torsional mode is equal to  $v_1^T=1.11v_t/dc=15$  cm<sup>-1</sup> and for particles with d=5.2 nm is equal to 11.9 cm<sup>-1</sup>, which is very close to the experimental values 14.2 cm<sup>-1</sup> and 12.1 cm<sup>-1</sup>.

### D. Comparison of theory to our experimental results

Let us now consider our experimental data. We measured the Raman scattering from Ge microcrystals in a GeO<sub>2</sub> matrix. In Fig. 3 the Raman spectra of a sample with different polarizations are shown. One can see that the peak maximum at polarized scattering (||||) is shifted to higher frequencies in comparison with the maximum at depolarized scattering ( $||\perp$ ), as was in the case of Raman scattering from CdS microcrystals in a GeO<sub>2</sub> matrix. The estimation has been done with the following values of parameters:

 $\rho_{\text{Ge}} = 5.33 \text{ g/cm}^3$ ,  $v_l = 5.25 \times 10^5 \text{ cm/sec}$ ,  $v_t = 3.25 \times 10^5 \text{ cm/sec}$  (Ref. 4),  $\rho_{\text{GeO}_2} = 3.6 \text{ g/cm}^3$ ,  $v_l = 3.43 \times 10^5 \text{ cm/sec}$ ,  $v_t = 2.14 \times 10^5 \text{ cm/sec}$  (Ref. 10).



FIG. 3. Polarization dependence of the reduced (by the Bose-Einstein occupation factor) low-frequency Raman spectra of Ge microcrystals embedded in a  $\text{GeO}_2$  matrix.

Coefficients in the determinants  $D_m^s$  and  $D_m^t$  are found to be equal to  $C_1=0.54$ ,  $C_2=0.82$ ,  $C_3=1.62$ ,  $C_4=1.6$ . The results of the estimation are shown in Fig. 2(c). The frequencies of the spheroidal modes with l=0 are equal to  $v_0^S = 0.8 v_t/dc$ ; i.e., they are not different from the vibrations of the particles with the free surface. This is due to the fact that the parameter  $C_1$  is farther from 1 than in the cases of CdS and Ag particles. It is known that the depolarized scattering is governed by spheroidal modes with l=2 or torsional modes with l=1. In an effort to determine which modes are involved in the scattering in the case in question we now turn to the low-frequency Raman spectra (Fig. 4) which were measured in Ref. 5 for the same sample with two wavelengths of exciting light, 514.5 and 647.1 nm. It turns out that the modes, located deeper under the surface of a microcrystal, on which the less absorbed light with  $\lambda = 647.1$  nm is scattered, have a higher vibrational frequency. Consequently, the spectra in Fig. 4 are due to torsional vibrational modes, because only



FIG. 4. The low-frequency depolarized Raman scattering spectra of Ge microcrystals of the size d = 9 nm for two wavelengths of the exciting light.

their frequency smoothly increases at increasing stiffness of the matrix and because they are located deeper under the surface of a microcrystal, as seen in Fig. 1. Evidently, a spread in frequency of the surface mode is due to the inhomogeneous form of the microcrystal-matrix interface. Just the surface modes take part in the Raman scattering, and it is seen from Fig. 2(c) that all the inner modes with  $n \ge 1$  are situated much farther in frequency than the difference of the frequencies observed in the experiment, both for the spheroidal and the torsional modes.

From all the above experiments it is clear that depolarized scattering is governed by surface torsional modes with l=1 which appear when the crystals are embedded in a matrix. Furthermore, from the calculations it follows that the influence of the matrix on the frequencies of the acoustic phonons, confined in the microcrystals, is significant even if Lame's constants  $\lambda$ ,  $\mu$  and the mass densities of the microcrystal and the matrix are quite different from each other, especially for n = 0 and 1 (see Fig. 2).

# **IV. CONCLUSION**

In conclusion, we note that the electron-phonon interaction with acoustic phonon modes in small microcrystals arises mainly through the deformation-potential coupling. The most dominant term can be described by  $E_d \cdot \operatorname{div} \vec{u}$ , where  $E_d$  is the deformation potential and  $\vec{u}$  is the lattice displacement vector.<sup>1</sup> If microcrystals have a perfect spherical form, torsional modes would not contribute to this coupling because of their transvere character. The deviation of the microcrystalline shape from the perfect spherical form may cause changes in symmetry and the Raman selection rules. The torsional modes become the mixed modes of the transverse and longitudinal nature and therefore it is possible to observe them by low-frequency Raman scattering.

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#### APPENDIX

The components of the matrix  $D_m^s$  which are used for determination of the eigenfrequencies  $\eta_{nl}^s$  of the spheroidal vibrational modes are

$$d_{11} = C_{3}^{2} j_{l}(\xi_{i}) - \frac{2}{\xi_{i}} \left[ \frac{l(l-1)}{\xi_{i}} j_{l}(\xi_{i}) + 2j_{l+1}(\xi_{i}) \right],$$
  

$$d_{12} = -C_{2}^{2} C_{3}^{2} \sqrt{\frac{\pi}{2\xi_{m}}} \left\{ C_{1}^{2} K_{l+1/2}(\xi_{m}) - \frac{2}{\xi_{m}} \left[ \frac{l(l-1)}{\xi_{m}} K_{l+1/2}(\xi_{m}) + 2K_{l+3/2}(\xi_{m}) \right] \right\},$$
  

$$d_{13} = 2C_{2}^{2} C_{3}^{2} l(l+1) \frac{1}{\eta_{i}} \left[ \frac{(l-1)}{\eta_{i}} j_{l}(\eta_{i}) - j_{l+1}(\eta_{i}) \right],$$

$$\begin{split} d_{14} &= -2C_2^2 \frac{1}{\eta_m} \sqrt{\frac{\pi}{2\eta_m}} \bigg[ \frac{(l-1)}{\eta_m} K_{l+1/2}(\eta_m) - K_{l+3/2}(\eta_m) \bigg], \\ d_{21} &= \frac{2}{\xi_i} \bigg[ \frac{(l-1)}{\xi_i} j_l(\xi_i) - j_{l+1}(\xi_i) \bigg], \\ d_{22} &= -2C_2^2 C_3^2 \frac{1}{\xi_m} \sqrt{\frac{\pi}{2\xi_m}} \bigg[ \frac{(l-1)}{\xi_m} K_{l+1/2}(\xi_m) - K_{l+3/2}(\xi_m) \bigg], \\ d_{23} &= -C_2^2 C_3^2 \bigg[ \bigg( \frac{2(l^2-1)}{\eta_i^2} - 1 \bigg) j_l(\eta_i) + \frac{2}{\eta_i} j_{l+1}(\eta_i) \bigg], \\ d_{24} &= C_2^2 \sqrt{\frac{\pi}{2\eta_m}} \bigg[ \bigg( \frac{2(l^2-1)}{\eta_m^2} - 1 \bigg) K_{l+1/2}(\eta_m) \\ &+ \frac{2}{\eta_m} K_{l+3/2}(\eta_m) \bigg], \end{split}$$

$$d_{13} = j_{l}(\xi_{i}), \quad d_{41} = 0,$$

$$d_{32} = -C_{4}^{2} \sqrt{\frac{\pi}{2\xi_{m}}} K_{l+1/2}(\xi_{m}), \quad d_{42} = 0,$$

$$d_{33} = 0, \quad d_{43} = C_{2}^{2} C_{3}^{2} j_{l}(\eta_{i}),$$

$$d_{34} = 0, \quad d_{44} = -\sqrt{\frac{\pi}{2\eta_{m}}} K_{l+1/2}(\eta_{m}),$$

$$\xi_{n,l} = \frac{\omega_{n,l} d}{2v_{l}}, \quad C_{3} = \{(\lambda_{i} + 2\mu_{i})/\mu_{i}\}^{1/2},$$

$$C_{4} = \{(\lambda_{m} + 2\mu_{m})/\mu_{m}\}^{1/2}.$$

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