Interface Phonons in Semiconductor Nanostructures with Quantum Dots

M. Yu. Ladanov\textsuperscript{a}, A. G. Milekhin\textsuperscript{a}, A. I. Toropov\textsuperscript{a}, A. K. Bakarov\textsuperscript{a}, A. K. Gutakovskii\textsuperscript{a}, D. A. Tenne\textsuperscript{b}, S. Schulze\textsuperscript{c}, and D. R. T. Zahn\textsuperscript{c}

\textsuperscript{a}Institute of Semiconductor Physics, Siberian Division, Russian Academy of Sciences, pr. Akademika Lavrent'eva 13, Novosibirsk, 630090 Russia
\textsuperscript{b}Department of Physics, Pennsylvania State University, 104 Davey Lab., University Park, PA 16802, USA
\textsuperscript{c}Institut für Physik, Technische Universität Chemnitz, D-09107, Chemnitz, Germany

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Abstract—The vibrational spectra of structures with InAs quantum dots in an AlGaAs matrix and AlAs quantum dots in an InAs matrix are investigated experimentally and theoretically. The Raman spectra exhibit features that correspond to transverse-optical (TO), longitudinal-optical (LO), and interface phonons. The frequencies of interface phonons in InAs and AlAs quantum dots and in an AlGaAs matrix with various concentrations of aluminum are calculated with the use of experimental values of transverse- and longitudinal-optical phonons in the approximation of a dielectric continuum. It is shown that the model of a dielectric continuum adequately describes the behavior of interface phonons in structures with quantum dots under the assumption that the quantum dots are spheroidal.

1. INTRODUCTION

Periodic semiconductor structures with self-organized quantum dots, which are characterized by unique electronic and optical properties, are one of the most challenging objects of research in semiconductor physics. These objects attract interest in view of the possibility to design, on the basis of these objects, new devices such as quantum transistors, high-speed memory elements, narrowband light-emitting diodes, heterojunction lasers, and infrared (IR) photodetectors [1–3].

Progress in the epitaxial growth technology has made it possible to produce quantum-dot structures with controllable properties on the basis of a series of materials (InAs/Ga(Al)As, In(Ga)As/InP \cite{4, 5}, Ge/Si, GaSb/InP, GaN/AlN) \cite{6–9}. The most thoroughly investigated system is InAs/Ga(Al)As; a large number of papers have been devoted to the study of its optical and electronic properties \cite{2}. However, despite the fact that the vibrational spectrum contains information about the structural properties (the size, dispersion of size, and the shape) of quantum dots \cite{10, 11} and mechanical stress in nanostructures \cite{12, 13}, the vibrational properties have been poorly studied even in this system. The most widespread methods for studying vibrational spectra are the Raman spectroscopy and the infrared (IR) spectroscopy. These methods are complementary because they use different selection rules; therefore, they allow one to study vibrational excitations of different types of symmetry. The Raman and IR spectroscopy have been applied to study optical phonons in stressed \cite{13, 14} and relaxed \cite{12, 15} quantum dots, in quantum dots of InGaAs solid solutions \cite{16}, and in a wetting layer \cite{17, 18}.

Earlier, a theoretical analysis of the spectrum of optical phonons in quantum dots was carried out within the model of valence-force fields \cite{19, 20} and in the approximation of a dielectric continuum \cite{15}. The model of valence-force fields is an empirical atomistic model and allows one to calculate the phonon frequencies in quantum dots consisting of a few thousand atoms. Calculations with the use of this model involve large arrays of data; this makes these calculations rather tedious. The approximation of a dielectric continuum is a macroscopic model and can rather easily be applied to the calculation of the frequencies of interface phonons localized near the interface between the materials of the quantum dots and the matrix \cite{21}.

The simplest model of a dielectric continuum deals with spherical quantum dots of one material embedded into the matrix of another material \cite{22, 23}. In this case, the eigenfrequencies are determined from the condition

\[
\frac{\epsilon_1(\omega_{lm})}{\epsilon_2(\omega_{lm})} = -1 - \frac{1}{l},
\]

where \(\epsilon_1\) and \(\epsilon_2\) are the dielectric functions of the quantum dots and the matrix, respectively; \(\omega_{lm}\) are the eigenfrequencies of interface phonons; and \(l\) is a quantum number of a phonon (\(l = 1, 2, \ldots\)).

As a rule, the shape of a real quantum dot is different from a sphere \cite{24, 25} (a truncated pyramid for a system of InAs quantum dots in a GaAs matrix \cite{26}, a
hemisphere for Ge quantum dots in a Si matrix [27], and an ellipsoid for AlAs quantum dots in an InAs matrix [28]). Therefore, the model of a dielectric continuum was further developed in [29, 30], where it was assumed that quantum dots are spheroidal. The reduction of the symmetry of quantum dots from spherical to spheroidal complicates the condition for the eigenfrequencies of interface phonons: these frequencies will now depend on two quantum numbers, \( l \) and \( m \).

In [30], the dielectric function of a matrix is considered that does not depend on frequency; this provides a unique set of interface modes whose frequencies lie between the frequencies of TO and LO phonons in quantum dots. Such an approach is justified if a model deals with quantum dots in a vitreous or an organic matrix.

In [29], it was assumed that the dielectric functions of both the quantum dots and the material of the matrix depend on frequency; unlike the dielectric-continuum model considered in [30], this yields two sets of interface modes. The first set, which refers to quantum dots, lies in the spectral range between TO and LO phonons in the material of quantum dots. The other set lies in the frequency range between the corresponding values of bulk phonons in the matrix material.

Despite the progress made in the theoretical description of interface phonons in spheroidal quantum dots, there is a lack of experimental research in interface phonons in structures with self-organized quantum dots [11, 13].

In this paper, we present the results of investigating interface phonons in structures with InAs and AlAs quantum dots by the methods of Raman spectroscopy and compare them with the data obtained by calculating the interface phonons in the approximation of a dielectric continuum.

2. THEORY

Let us write out the basic equations necessary for the analysis of the dielectric-continuum approximation in polar materials [30, 31]. The Born–Huang equation of motion can be represented as follows:

\[
\dot{\mathbf{w}} = -\omega_{TO}^2 \mathbf{w} + \frac{\varepsilon_0 - \varepsilon_\infty}{\mu} \frac{\omega_{TO}^2}{4\pi} \mathbf{E},
\]

where the polarization \( \mathbf{P} \) can be expressed as

\[
\mathbf{P} = \frac{\varepsilon_0 - \varepsilon_\infty}{\mu} \frac{\omega_{TO}^2}{4\pi} \mathbf{w} + \frac{\varepsilon_\infty - 1}{4\pi} \mathbf{E}.
\]

Here, \( \mathbf{w} = \sqrt{N/\mu} \mathbf{u} \), where \( \mathbf{u} \) is a relative displacement between a pair of ions with reduced mass \( \mu \) in a crystal with concentration \( N \), \( \mathbf{E} \) is the electric field, \( \omega_{TO} \) and \( \omega_{LO} \) are the frequencies of transverse- and longitudinal-optical phonons, and \( \varepsilon_0 \) (\( \varepsilon_\infty \)) is the static (high-frequency) dielectric constant of a polar material. In addition, we assume that the Lidden–Sacks–Teller relation \( \omega_{LO}^2 / \omega_{TO}^2 = \varepsilon_0 / \varepsilon_\infty \) holds.

For the electric field to satisfy the Maxwell equations, it is necessary that the electric induction

\[
\mathbf{D} = \varepsilon(\omega) \mathbf{E} = \mathbf{E} + 4\pi \mathbf{P}
\]

should satisfy the Gauss equation

\[
\mathbf{\nabla} \cdot \mathbf{D} = 0.
\]

Using the relation

\[
\mathbf{E} = -\mathbf{\nabla} \phi,
\]

we can write out the basic equation of dielectric approximation:

\[
\varepsilon(\omega) \mathbf{\nabla}^2 \phi = 0.
\]

It is assumed that the time dependence of all the quantities introduced above is harmonic: \( f(t) \propto \exp(-i\omega t) \). In the absence of damping, the frequency-dependent dielectric function \( \varepsilon(\omega) \) of a polar material is defined by

\[
\varepsilon(\omega) = \varepsilon_\infty - \frac{\omega_{LO}^2 - \omega^2}{\omega_{TO}^2 - \omega^2}.
\]

Interface phonons are directly related to the electric potential, which must satisfy the Laplace equation \( \mathbf{\nabla}^2 \phi = 0 \). Therefore, one of the possible solutions to Eq. (4) is \( \varepsilon(\omega) \neq 0 \) for \( \omega \neq \omega_{LO} \). The boundary condition at the interface \( S \) between two media, the continuity of the normal components of \( \mathbf{D} \), is expressed as

\[
\varepsilon_1 \left[ \frac{\partial \phi_1}{\partial n} \right]_S = \varepsilon_2 \left[ \frac{\partial \phi_2}{\partial n} \right]_S.
\]

Since the object of our study are interface phonons in spheroidal quantum dots, it is convenient to pass from Cartesian coordinates to spheroidal (prolate and oblate) coordinate systems [30].

The prolate system of coordinates \( \xi, \eta, \phi \) is used for calculating the frequencies of interface phonons for prolate quantum dots and is expressed in terms of Cartesian coordinates as follows:

\[
x = b \sqrt{\left( \xi^2 - 1 \right) \left( 1 - \eta^2 \right)} \cos \phi, \\
y = b \sqrt{\left( \xi^2 - 1 \right) \left( 1 - \eta^2 \right)} \sin \phi, \\
z = b \xi \eta,
\]

whereas the oblate system of coordinates is convenient for determining the frequencies of interface phonons in oblate quantum dots:

\[
x = b \sqrt{\left( \xi^2 + 1 \right) \left( 1 - \eta^2 \right)} \cos \phi, \\
y = b \sqrt{\left( \xi^2 + 1 \right) \left( 1 - \eta^2 \right)} \sin \phi, \\
z = b \xi \eta,
\]