1. INTRODUCTION

The effects associated with the interaction between translational and rotational degrees of freedom in molecular crystals and solid solutions containing molecular components manifest themselves in many physical properties of these solids [1]. Examples are provided by the low-frequency anomalies of the transverse phonon modes in molecular and ionic–molecular lattices, the specific features in the temperature dependences of the elastic moduli for a number of molecular crystals, structural distortion of the orientationally ordered phases due to phonon–libron interaction, etc. (see [1] and references therein). The aforementioned effects are best understood in classical solids. However, there are a number of works devoted to the investigation of these phenomena in cryocrystals [2, 3]. Apart from the results regarding the thermodynamics of cryosystems in the presence of phonon–libron interaction, etc. (see [1] and references therein), there are a number of works devoted to the investigation of these phenomena in molecular cryocrystals [2, 3].

Two-dimensional cryosolutions involving molecular impurities are substantially more complicated systems. These systems are prepared by applying atomic–molecular mixtures on substrates (most frequently represented by graphite and some metals) [16–28]. In this context, phonons and librions are the sole possible types of collective excitations. Moreover, the conditions favorable for observing specific quantum effects that have no analogs in classical solids can be achieved at low temperatures.

Solid solutions of homonuclear and heteronuclear molecules in atomic matrices composed of solidified rare gases form a particular group of objects whose properties can be substantially affected by translational–rotational interaction. As was shown earlier in [10–15], the rotational constant $B$ of an impurity and the crystal field constant $\tilde{\mathcal{H}}$ in three-dimensional solutions prove to be extremely sensitive to the presence of translational modes in the lattice. In particular, the interaction between a molecule rotator and short-wavelength phonons leads to the fact that the values of $B$ and $\tilde{\mathcal{H}}$ for an impurity in the atomic cryomatrix differ significantly from those for a free molecule [13–15]. Consideration of this circumstance made it possible to interpret successfully the experimental data on the heat capacity of Ar(Kr)–N$_2$ (O$_2$) solutions [12, 14, 15].

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As a consequence, the dynamics of the impurity molecule in the two-dimensional solution appears to be substantially more complex than that in the three-dimensional system. This can appreciably affect the behavior of all physical characteristics. At present, the above problem has not been adequately studied. The purpose of the present work is to fill this gap.

2. FORMULATION OF THE PROBLEM

Let us consider the problem of motion of a diatomic homonuclear molecule in a two-dimensional close-packed monoatomic matrix (a solidified rare gas) located on a substrate composed of atoms of the same compound. The interaction between the impurity and the environment will be included in the atom–atom potential approximation [29]. In the system under consideration, the interactions of the impurity with the substrate can be described by potentials of the same type (for example, the Lenard-Jones potential). Note that, in the general case, the parameters of the potentials corresponding to the impurity–matrix and impurity–substrate interactions should be treated as being different.

It is assumed that the origin of the coordinates is located at a lattice site in which a matrix atom is replaced by a diatomic homonuclear impurity molecule with mass $M$ and internuclear distance $2d$. The configuration of the system is displayed in the figure. The matrix and the substrate represent two-dimensional atomic layers with a triangular lattice. The $0Z$ axis of the Cartesian coordinate system is perpendicular to the atomic layers, and the $0X$ and $0Y$ axes are oriented along the matrix plane. The interatomic distances in both the matrix and the substrate are equal to $R_1$, and the distance between the layer and substrate is defined as $c = \gamma R_1$ (for a perfect close packing of hard spheres, $\gamma = \sqrt{2/3}$). Since the parameters of the interaction of matrix atoms with each other and with the impurity molecule are different, the equilibrium position $O_c$ of the center of inertia of the impurity does not coincide with the layer plane $z = 0$. From simple symmetrical considerations, the point $O_c$ lies on the $0Z$ axis at a certain distance $z_0$ from the origin of the coordinates.

The complete Hamiltonian of the impurity–matrix–substrate system has the form

$$H = \frac{\mathbf{p}^2}{2M} + B\mathbf{L}^2 + U. \quad (1)$$

Hereafter, we will use a system of units in which the Planck and Boltzmann constants are equal to unity. The first term in formula (1) is the translational kinetic energy for the center of inertia of the molecule. The quantity $\mathbf{p} = -i\partial/\partial \mathbf{u}$ is the momentum of the impurity, and $\mathbf{u}$ is the displacement of the center of inertia of the impurity from the equilibrium position $O_c$. The second term in Hamiltonian (1) is the rotational kinetic energy for the impurity molecule. In this case, $B = 1/(2J)$ is the rotational constant, $J = Md^2$ is the moment of inertia of the impurity, and $\mathbf{L}^2$ is the square of the angular momentum of the impurity, which can be represented in the form

$$\mathbf{L}^2 = \left[ \frac{1}{\sin \phi} \frac{\partial}{\partial \phi} \left( \sin \phi \frac{\partial}{\partial \phi} \right) + \frac{1}{\sin^2 \phi} \frac{\partial^2}{\partial \phi^2} \right]. \quad (2)$$

Here, the angles $\phi$ and $\varphi$, which specify the orientation of the rotator, are determined as follows: the $\phi$ angle is measured from the $0Z$ axis and the $\varphi$ angle is reckoned from the $0X$ axis.