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Electron-phonon interaction, phonon and electronic structures of layered electride Ca_2N

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The phonon and electronic properties, the Eliashberg function and the temperature dependence of resistance of electride Ca_2N are investigated by the DFT-LDA (density functional theory in local density approximation) plane-wave method. The phonon dispersion, the partial phonon density of states and the atomic eigenvectors of zero-center phonons are studied. The electronic band dispersion and partial density of states conclude that Ca_2N is a metal and the Ca 3p, 4s and N 2p orbitals are hybridized. For the analysis of an electron-phonon interaction and its contribution of the Eliashberg function to resistance was calculated and a temperature dependence of resistance due to electron-phonon interaction was found.

There is considerable interest in layered electrides in bulk and monolayer forms in which *playing anionic role* electrons form two-dimensional planes separated from positively charged layers of ions [1–6]. In view of their promising properties such as high electrical conductivity, low work function, and significant catalytic activity in their ideal form, electrides are perspective for use in next-generation electronics.

Sub-nitride Ca_2N belongs to this new class of layered-structure electrides with the two-dimensional delocalized layers of electrons [7, 8]. The rhombohedral unit cell of Ca_2N contains more electrons than it is expected from the simple electron counting rules. It is supposed that excess electrons are localized between positively charged layers $[Ca_2N]^+$. The physical properties of Ca_2N were studied by photoelectron spectroscopy [7, 9], optical reflectance spectroscopy [7], electrical conductivity [7, 10], by study of magnetic susceptibility [10] and magnetoresistance [7]. It was found that single crystal Ca_2N exhibits metallic transport with resistivity, which is smaller than that of pure Ca metal [7]. The temperature dependence of resistivity indicated that the electron-electron interaction could be much stronger than the electron-phonon interaction even in the high-temperature region [7].

In this Letter we calculate the Eliashberg function of Ca_2N from first principles (DFT) to estimate the electron-phonon interaction (EPI) and its contribution to resistance as well as a temperature dependence of resistance caused EPI.

We start our calculations from the study the phonon and electronic properties of the Ca_2N crystal in order to compare with calculations [11, 12] of these properties in the isolated layer Ca_2N and to obtain the density of phonon states $F(\omega)$ that is necessary to compute EPI. Besides, in distinct from previous calculations [11, 12], we had to use the relativistic pseudopotentials [13, 14] in first-principles calculations of EPI. The frequencies of phonon branches have been calculated within density perturbation theory [15]. There was no an energy gap between acoustic and optic branches in the phonon dispersion. If the main contribution of the N atoms in phonon density $F(\omega)$ locates above $285\,\mathrm{cm}^{-1}$, the Ca atom gives a contribution at lower frequencies except for acoustic modes in which a participation of both atoms is noticeable. We have found, the electron density of states at the Fermi level is finite, that is, Ca_2N is a metal.

The role of different phonon branches in electron-phonon interactions can be characterized by the spectral Eliashberg function $\alpha^2F(\omega)$ [16, 17] where α^2 is the squared effective EPI. The calculations of $\alpha^2F(\omega)$ were performed by the method [18]. The main peaks in phonon density $F(\omega)$ are seen also in function $\alpha^2F(\omega)$, confirming a contribution of almost all phonons to the Eliashberg function in Ca_2N. We calculate the resistance only due to electron-phonon interactions. In this

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case the temperature dependence of electrical resistivity can be calculated in the lowest-order variational approximation [18] using the calculated function $\alpha^2 F(\omega)$, density of the electronic states at the Fermi level and average square of the Fermi velocity for electrons. The calculated resistance ρ of Ca_2N turned out to be highly anisotropic, and it is visibly higher along the axis z. This result is consistent with experimental estimates. The small value of ρ_x is in agreement with idea of presence of a two-dimensional layer of electrons between positively charged layers $[Ca_2N]^+$ [7, 8] and with our calculated electronic structure, from which it follows that Ca_2N should have a metallic character along the plane of the layers.

In summary, we have presented a first-principles study of phonon and electronic structures and resistance of electride Ca_2N at the LDA level taking into account semi-core electrons for the Ca atom and using the plane-wave pseudopotential method. The phonon properties such as the phonon dispersion, the partial phonon density of states and the eigenvectors of zone-center optic modes are investigated. All phonons make a significant contribution to electron-phonon interactions in Ca_2N. The calculated resistance of Ca_2N turned out to be highly anisotropic, and it is visibly lower along the plane of the layers.

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