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Nonadiabatic Interaction between Electrons and Jahn-Teller Distortions in a Silicon Vacancy

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Abstract

Recently, Goto *et al.* have discovered anomalous elastic softening due to vacancy in crystalline silicon at very low temperature. To investigate the quantum state of the silicon vacancy, we introduce a cluster model which includes the electron-lattice coupling between electrons and Jahn-Teller distortions together with the Coulomb interaction between electrons. By using the numerical diagonalization method, we take into account of both the nonadiabatic and the correlation effects. We find that the 3-fold orbital degeneracy in the ground state of a silicon vacancy is stable against the Jahn-Teller distortions because of the strong quantum fluctuation due to the nonadiabatic coupling between electrons and Jahn-Teller distortions. The obtained result is a striking contrast to the previous theoretical results within the adiabatic approximation and is consistent with the newly observed experiment.

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Lattice defect in crystalline silicon is one of the most influential factors to determine the quality of semiconductor devices. Although a void, which is a collection of vacancies, is observable, it is difficult to observe a single vacancy itself. Recently, Goto *et al.* [1] have discovered an anomalous elastic softening in pure crystalline silicon at low temperature below 20K down to 20mK. Remarkably, the softening is independent of the external magnetic field up to 16T. Then, the elastic softening is attributed to the 3-fold orbital degeneracy in the ground state of a silicon vacancy.

In early theoretical studies, Schlüter *et al.* [2,3] showed that the 3-fold orbital degeneracy in the ground state of a silicon vacancy is removed due to the Jahn-Teller distortions within the adiabatic approximation. It seems to be inconsistent with the low-temperature elastic softening. However, the nonadiabatic effect of the Jahn-Teller distortions, which is considered to play crucial role at low temperature, was not considered there. With the advent of the new findings, theoretical studies of the silicon vacancy state including the nonadiabatic effect of the Jahn-Teller distortions is highly desirable.

In this paper, we investigate the silicon vacancy state, paying attention to the effect of the nonadiabatic Jahn-Teller distortions strongly coupled to electrons via the electron-phonon coupling, together with the correlation effect due to the electron-electron Coulomb interaction. For this purpose, we introduce a cluster model as a simple extension to the previous model derived by Schlüter *et al.* for a silicon vacancy on the basis of the first-principle calculation [2,3].

Our model Hamiltonian includes 6 orbitals, 4 orbitals ($i = 1-4$) of dangling bonds in the nearest neighbor atoms of the vacancy and 2 orbitals from the valence band ($i = 5$) and the conduction band ($i = 6$), respectively:

$$\mathcal{H} = \sum_{i=1}^{6}\sum_{j=1}^{6}\sum_{\sigma=\pm1} t_{ij}a_{i\sigma}^{\dagger}a_{j\sigma} + U\sum_{i=1}^{4} a_{i\uparrow}^{\dagger}a_{i\uparrow}a_{i\downarrow}^{\dagger}a_{i\downarrow} + \frac{1}{2M_E}(P_{Ex}^2 + P_{Ey}^2 + P_{Ez}^2) + \frac{1}{2M_B}P_B^2 + \frac{1}{2}K_E(Q_{Ex}^2 + Q_{Ey}^2 + Q_{Ez}^2) + \frac{1}{2}K_B Q_B^2 \quad (1)$$

where $a_{i\sigma}^{\dagger}$ is the creation operator for an electron at site i with spin σ and U is the on-site Coulomb interaction. Q_{Ex}, Q_{Ey}, Q_{Ez} (Q_B) are tetragonal (breathing) mode distortions with the effective mass M_E (M_B) together with the

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spring constant K_E (K_B), respectively. In eq.(1), transfer integrals t_{ij} are explicitly given by

$$
\begin{aligned}
t_{i,i} &= \epsilon \qquad \text{for} \quad i \in \{1,2,3,4\}, \\
t_{5,5} &= \epsilon_C, \quad t_{6,6} = \epsilon_V, \quad t_{5,6} = 0, \\
t_{1,2} &= t_{3,4} = -t - \frac{1}{\sqrt{3}} g_E Q_{Ex} - g_B Q_B, \\
t_{1,3} &= t_{2,4} = -t - \frac{1}{\sqrt{3}} g_E Q_{Ey} - g_B Q_B, \\
t_{1,4} &= t_{2,3} = -t - \frac{1}{\sqrt{3}} g_E Q_{Ez} - g_B Q_B, \\
t_{i,5} &= t_C, \quad t_{i,6} = t_V \qquad \text{for} \quad i \in \{1,2,3,4\},
\end{aligned}
\tag{2}
$$

where g_E and g_B are the electron-lattice coupling constants for the tetragonal mode and the breathing mode, respectively. The parameters of this model are determined so as to reproduce the previous theoretical results [2,3] within the adiabatic approximation for the distortions together with the mean-field approximation for the Coulomb interaction.

Here, we replace the distortion by the phonon operator, $Q_\nu = \sqrt{\hbar/2M_\nu\omega_\nu}(b_\nu + b_\nu^\dagger)$, with $\omega_\nu = \sqrt{K_\nu/M_\nu}$, for each mode ν. Then, we take into account of both effects of the nonadiabatic electron-lattice coupling and the electron correlation by using the numerical diagnalization with the standard Lanczos algorithm for the Hamiltonian eq.(1) with eq.(2), where the cutoff of the phonon number is set to be 7 for each mode.

In Fig. 1, we plot the energy difference between the various occupancy states of a silicon vacancy, where the electron occupation number in the 4 orbitals of the dangling bonds are 4 in the V^0 state, 3 in the V^+ state and 2 in the V^{++} state, respectively. The solid lines represent boundaries separating the different occupancy states. We note that the V^+ state is always unstable, in contrast to the previous mean-field results [2,3] where the V^+ state becomes stable for large U as an artifact of the approximation.

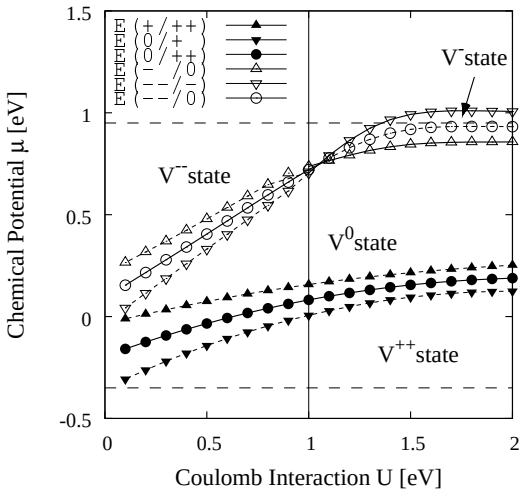

Fig. 1. Stable occupancy state of a silicon vacancy as a function of U and μ. The vertical solid line shows the parameter corresponding to the Schlüter's result. The upper and the lower dashed horizontal lines represent ϵ_C and ϵ_V, respectively.

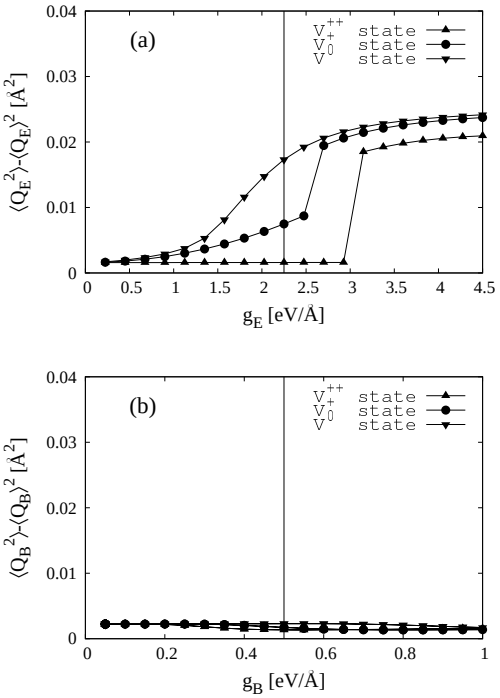

Fig. 2. Mean square distortions for the tetragonal mode vs. g_E (a) and for the breathing mode vs. g_B (b).

Fig. 2 shows the lattice fluctuations as functions of the electron-lattice coupling. When g_E increases, the tetragonal mode fluctuations for the V^0 and V^+ states increase, because the both states have 3-fold orbital degeneracy and the electrons strongly couple to the tetragonal distortions due to dynamical Jahn-Teller effects. On the other hand, in the V^{++} state without orbital degeneracy, the lattice fluctuation is almost constant for small g_E. In the V^+ and V^{++} states, a remarkable transition to the V^0-like state is observed in the strong coupling regime. As shown in Fig. 2(b), the breathing mode fluctuations are almost independent of g_B for all occupancy states. We infer from our results that the dynamical Jahn-Teller effect in the V^0 state plays an important role for the low-temperature elastic softening.

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