Reply to "Comment on 'Suppression of inelastic bound-state resonance effects by the dimensionality of an atom-surface scattering event' "

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We present a detailed study of the bound-state phonon-mediated resonances in atom-surface scattering that extends our earlier work [Šiber and Gumhalter, Phys. Rev. B **71**, 081401 (2005)]. Previous results obtained within the restricted Fock space coupled-channel algorithm are reconsidered in view of the enlargement and refinement of the basis of quantum states used in the calculation. The convergence demonstrated in this procedure reinforces the usefulness of the developed algorithm.

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We recently demonstrated the application of the coupled-channel method to the problem of scattering of inert gas atoms from surface phonons.[1] The application represents an extension of our earlier work reported in Refs. 2 and 3 and allows for a consistent treatment of lateral momentum transfer in multiphonon-assisted bound-state resonant atom-surface scattering. The proposed algorithm, which we termed the restricted Fock-space coupled channel (RFCC) method, enabled us to investigate the effects of bound-state resonances on the scattering of thermal energy He atoms from nondispersive surface phonons, and in particular to assess whether in the full three-dimensional (3D) treatment the strengths of inelastic scattering resonances should appear weaker than in the earlier proposed one-dimensional (1D) treatments of the same problem.[5,6] Application of the developed algorithm to the benchmark model describing low-energy He atom scattering from vertically polarized Einstein phonons (of frequency $\hbar\omega_Q \simeq 2.7$ meV) in Xe monolayers on the Cu(111) surface[4] showed that this indeed was the case (cf. Fig. 2 of Ref. 1).

The results of application of the developed algorithm convey two important messages concerning the theoretical studies of inelastic resonant scattering.

(i) Efficient numerical treatment of inelastic bound-state resonance effects in a multidimensional phonon phase space is feasible within a framework that includes the features of earlier multiphonon theories[7] [as demonstrated by comparison of the exponentiated Born approximation (EBA) with the RFCC results in Figs. 1 and 2 of Ref. 1].

(ii) The relative intensities of inelastic bound-state resonances are suppressed (albeit not completely eliminated) in going from 1D to 3D (cf. Fig. 2 of Ref. 1). As we understand, the author of the proceeding Comment agrees with us on both points (i) and (ii). Specifically, according to our 3D calculations applied to the He→Xe/Cu(111) collision system, the phonon-assisted bound-state resonance effects can induce ~4% change in the relative elastic scattering intensity described by the so-called Debye-Waller factor P_0, whereas in 1D this effect is around 40%. This indicates that under these circumstances the inelastic bound-state resonance effects would be difficult to observe in the He →Xe/Cu(111) scattering experiments.

In a Comment[8] on our work Brenig argues that the bound-state phonon-mediated resonances we discerned in the calculated 3D total elastic scattering probability $P_0(E_i)$ as a function of the incident He atom energy E_i are at least a factor of 2 smaller than the ones he could obtain in a somewhat different 3D treatment. In his opinion there are some shortcomings in our approach for which he suggests improvements in the domain of obtaining better numerical accuracy. Application of a modified CC treatment to the same collision system led him to the conclusion that under favorable circumstances it might be possible to observe the discussed resonance effects experimentally. In the following we shall demonstrate that a reconsideration of the same problem in which our method is implemented with increased numerical accuracy leads to the same conclusions as reached in Ref. 1.

The shortcomings that Brenig associates with our method can be explained simply by the insufficient density of points in the set of incident projectile energies that were sampled in Ref. 1, as Brenig also acknowledges.[8] Since the calculated resonances are rather narrow (about 0.03 meV; see Fig. 1 below and Fig. 1 of Brenig's comment), a sufficiently dense set of projectile incident energies needs to be considered to provide proper resonance sampling. To show this we have repeated the numerical calculations of $P_0(E_i)$ with an increased number of calculated points per incident energy interval. Thus, the spacing of the energy mesh in the present calculation is 0.01 meV, i.e., about the same as in the calculation of Brenig[8] and over the same incident energy interval. The results of these calculations are presented in Fig. 1 (note in passing that in Ref. 1 the Debye-Waller factor was investigated over much larger energy intervals).

It is apparent that the resonance peaks appearing in the Debye-Waller factor $P_0(E_i)$ recalculated by using a denser energy mesh reach the same heights (on the average) as those in Fig. 1 of the Comment.[8] A closer inspection of Fig. 1(b) reveals that the total number and positions of resonances obtained in the present calculation are the same as in the earlier one,[1] only the relative heights of resonance peaks over the non-resonant structure are, on the average, a factor of $\leqslant 2$ larger (i.e., in total ~2% larger) due to a finer sampling of the incident energy parameter space (Brenig's quotation of a disagreement of a factor of 4 is exaggerated). On the other hand, one should not be surprised that the exact positions of resonances as well as their number are not the same in the present calculations and in those performed by Brenig.[8] These characteristics strongly depend on the choice

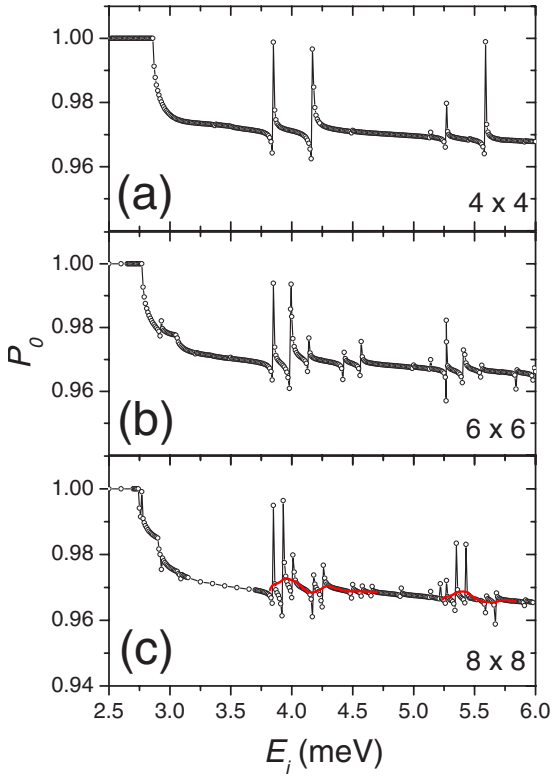

FIG. 1. (Color online) Debye-Waller factor (P_0) for model He $\rightarrow$Xe/Cu system as a function of the energy E_i of a He atom at normal incidence to a cold surface. (a), (b), and (c) display the results of the calculations with 4×4, 6×6, and 8×8 meshes in **Q** space, respectively. Calculated points are denoted by open circles, thin full lines are guides to the eye. The thick full line in (c) denotes a convolution of $P_0(E_i)$ with a Gaussian of half-width σ =0.04 meV which accounts for the effects brought about by the broadening mechanisms.

of mesh in the phonon lateral momentum (**Q**) space and hence they should not be assigned other content because different discrete **Q** meshes reveal different resonant signatures. This is also illustrated in Fig. 1 where one can see how the resonant signatures change with the sampling of the **Q** space. Of course, in the limit of a very dense **Q** mesh the individual resonances would merge into a continuum of resonances and the details of the **Q**-space sampling should become irrelevant. Therefore, the comparison of Brenig's $7\times5=35$ and our $6\times6=36$ **Q**-point calculations that would go into mesh-induced details of the computed spectra is neither meaningful nor useful and may even be misleading, as we shall now explain on the basis of our recalculated results.

In the results presented in the Comment[8] Brenig obtained a larger number of resonances since his choice of mesh, which is asymmetric (7×5 instead of 6×6 considered in Ref. 1) yields a larger number of different absolute values $|\mathbf{Q}|=Q$ of the phonon wave vector **Q**. Thus, Brenig's asymmetric set of **Q** values is less "degenerate" with respect to Q than our symmetric set, and as the quantity Q enters the momentum and energy conservation laws that determine resonance positions for normal projectile incidence, Brenig obtains a correspondingly larger number of resonances. However, this is achieved in a rather artificial fashion since Brenig's choice of the **Q** mesh introduces an unphysical asymmetry in the treatment of phonon wave vectors over the central part of the surface phonon Brillouin zone (seven spacings in, say, the x direction and five spacings in the y direction). Moreover, some of the resonances Brenig obtains are also related to inclusion of the states with two phonons having the same lateral momentum in the basis set of states employed in the calculation, which we excluded.[1] The argument invoked by Brenig to support this particular choice of the mesh was "to take into account the $\mathbf{Q}=\mathbf{0}$ momenta" that yield maximum excitation probability. However, this is also properly taken into account by our calculation, and without introduction of any artificial asymmetry in the surface Brillouin zone, as can be easily seen from Fig. 1 in the following way. Since $\mathbf{Q}=\mathbf{0}$ excitations not only contribute to the resonant scattering, but also to nonresonant inelastic scattering from phonons, any poor sampling of the **Q** space around the $\mathbf{Q}=\mathbf{0}$ point would be visible in the total inelastic scattering probability in the nonresonant region of incident energies, i.e., it would be smaller and consequently P_0 would be larger than in the calculation that properly accounts for the $\mathbf{Q}=\mathbf{0}$ excitations. For example, for $E_i=3.55$ meV (a nonresonant region) we obtain $P_0=0.970\,05$ (6×6 calculation) which is practically the same as Brenig obtained in his Fig. 1. The convergence of the nonresonant elastic scattering probability P_0 can also be seen from our Fig. 1 where one can note that already the 4×4 calculation nicely accounts for the value of the Debye-Waller factor in the nonresonant region ($P_0=0.972\,08$ for $E_i=3.55$ meV), while 6×6 ($P_0=0.970\,05$) and 8×8 calculations ($P_0=0.969\,94$) for the same E_i obviously exhibit excellent convergence. The convergence of Brenig's calculation with respect to the enlargement of the channel basis that he advocates cannot be estimated from the results presented in the Comment as these were obtained for a fixed **Q** mesh.

The calculated resonant structures in all three panels of Fig. 1 are very narrow. Hence, any broadening mechanism that may smear out their intensity would make the peaks less pronounced. This is illustrated in Fig. 1(c) in which we also show a convolution of the Debye-Waller factor $P_0(E_i)$ with a Gaussian of half-width $\sigma=0.04$ meV intended to conform with the procedure that was also invoked in the Comment. In a convolution the resonance effects show up as a $\sim1\%$ variation of P_0 in the resonant regions (humps centered around 4 and 5.5 meV). Here it should be noted that we have applied the convolution procedure to $P_0(E_i)$ which is a measurable quantity (obtained as the relative intensity of the elastically scattered beam[9]). This is in contrast to the Comment where the convolution procedure was applied to the angular integrated inelastic scattering intensity $1-P_0(E_i)$ which is not directly accessible in current experiments. That is, its measurement would require energy-resolved detection of all the particles inelastically scattered in the half-space above the surface, which is not feasible. Instead, it may be attempted to detect the above discussed resonance effects either as a discernible variation (increase) of the relative intensity of the elastically scattered beam in the region around E_i ~5.5 meV (which is at the borderline for production of monochromatic incident beams of He monomers[12]), or rather

in the energy- and angular-resolved (**Q**-resolved) measurements, where they may appear as sharper features in the same energy interval under the condition of high angular resolution.

Concerning a certain doubt that Brenig has cast[8] on the extensively used log-derivative method employed in our calculations, we merely want to point out that the same method has been previously implemented in very different contexts including studies of atom diffraction from and adsorption on corrugated surfaces,[3] rotationally inelastic scattering of molecules from surfaces,[10] and adsorption of He atoms in carbon nanotube materials.[11] Never have any problems been encountered in these implementations and we are confident that there are none in the present case either.

Last, although this is not directly related to the results of our work,[1] we may agree with Brenig's suggestion that the systems characterized by the parameters he uses to model the $Ne \rightarrow Xe/Cu$ collisions may be better candidates for discerning the bound-state phonon resonances than the discussed case of $He \rightarrow Xe/Cu$. However, from the computational point of view the large inelasticity in such collisions would most probably require an extension of the Fock space[1] beyond the two-phonon excitations and a corresponding check of convergence of calculations with the thus extended Fock space, as was done in Fig. 1 of Ref. 1. On the other hand, from the experimental point of view the observability of the resonance effects may in this case be hindered by the clustering of atomic Ne in such low-incident-energy beams.

In conclusion, we have demonstrated by simply doubling the phonon wave vector mesh density and energy resolution in our calculations that the algorithm developed in Ref. 1 does not suffer from the shortcomings pointed out in the preceding Comment.[8] The only difference occurring in comparisons of calculations presented in Refs. 1 and 8 arises from the procedure by which the set of projectile incident energies and values of exchanged phonon wave vectors are sampled in the numerical evaluation of the scattering probabilities. However, this does not in any way invalidate the main conclusion of Ref. 1 that the phonon-assisted bound-state resonances become much less pronounced in the full three-dimensional (3D) treatment of atom-surface scattering than in the 1D one, giving rise to only a few percent effect in the Debye-Waller factor for scattering of thermal energy He atoms from monolayers of Xe atoms adsorbed on Cu(111) surfaces.

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