

Mode-dependent energy transmission across nanotube junctions calculated with a lattice dynamics approach

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A generalized scattering boundary equation method (SBM) is proposed for the calculation of the vibrational amplitude transmission across solid junctions. An energy transmission expression with relation to atomic masses, lattice constants, and group velocities is elucidated. The phonon transmission is computed by the SBM for an (11,0)–(8,0) nanotube junction. The strongly mode-dependent energy transmission observed can be ascribed to the mismatch in symmetry of the vibrational motions, indicating its possibly important role in thermal transport in nanotubes.

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I. INTRODUCTION

Thermal transport in carbon nanotubes¹ has recently received much attention both theoretically and experimentally because of its unusually high thermal conductivity and possible engineering applications. Compared with three-dimensional bulk materials, the nanotubes have four acoustic-phonon branches and many optical-phonon branches,² which can be activated for thermal transport at a moderate temperature. However, the existence of interfaces and junctions among nanotubes or between nanotubes and their substrates is inevitable and will play an important role in nanoscale thermal transport.³ The different polarized phonon modes will be scattered by interfaces or junctions. An understanding of the scattering mechanism of each polarized phonon mode across interfaces or junctions is highly desired. Here we consider the energy transmission for different phonon modes across a nanotube junction within the atomistic lattice dynamic (LD) approach.

A scattering boundary method within the LD approach was first proposed in Refs. 4 and 5 for the calculation of energy transmission across an epitaxial interface with identical atomic masses on each side and with equal lattice constants. Nevertheless, for complex interfaces or junctions, a more general method is required to consider phonon scattering by junctions. In addition, the atomic masses and lattice constants on each side are usually different in realistic materials. So we need a general energy transmission expression within the scattering boundary lattice dynamic approach to incorporate these factors.

This work aims to (i) generalize the scattering boundary method for the epitaxial interface in Refs. 4 and 5 to the scattering boundary condition equation method (SBM) for the consideration of more complex junction atoms, and provide an energy transmission expression for an arbitrary lattice lead, elucidating its relation with atomic mass, lattice constant, and group velocity; (ii) study the energy transmission for different phonon modes across an (11,0)–(8,0) nanotube junction by the SBM to show the symmetrical mode-dependent phonon scattering across nanotube junctions.

We proceed as follows. In Sec. II, we will propose the SBM to calculate amplitude transmission across solid junctions and give a more general energy transmission expression within lattice dynamic approach. We will also clarify the relation of the SBM with other lattice dynamic approach.⁶ In Sec. III, we present the numerical result of transmission across a carbon-nanotube junction calculated by the SBM and expound the symmetrically mode-dependent phonon scattering across nanotube junctions.

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II. METHOD AND FORMULA

A. Scattering boundary condition equation

We consider the problem of energy transmission across a solid junction from the lattice dynamic approach by the scattering boundary method. The system considered comprises three parts: two ideal leads and a junction part in the center. Under the harmonic approximation, the whole system has the following linear dynamic equation of motion⁷ at a given frequency ω for each atom,

$$-m_i\omega^2\mathbf{u}_i + \sum_j \mathbf{K}_{i,j} \cdot \mathbf{u}_j = 0, \quad (1)$$

where m_i denotes the atomic mass for each atom and $\mathbf{u}_i$ is the oscillation amplitude. The force constants $\mathbf{K}_{i,j}$ can be found through the second-order derivatives of the potential. We will discuss the solution of Eq. (1) for atoms in the *whole* system considered to obtain the energy transmission across the solid junction. When the reservoirs are present on each side (lead), arbitrary lattice waves are excited on each side and then are scattered by the junction.

Plane wave solutions on the lead. Supposing that the junction part is relatively small in comparison with the two leads, the two leads can be assumed as *semi-infinite* lead model. This is often justified in realistic material structure because the interface or the junction part is usually small. Apart from its extensive application in modeling of electron transport,¹⁰ this semi-infinite lead model has been used for thermal transport of the epitaxial interface^{4,5} and thermal transport in nanowires.⁶ If the two semi-infinite leads are perfect crystals, the vibration of atoms in the leads can be further written as a combination of their plane wave modes, utilizing the translational periodicity in the leads. The eigen-

mode displacement $\mathbf{u}_{l,i,n}(\omega, \mathbf{q})$ for the atom (l, i) in the lead can be written as⁷

$$\mathbf{u}_{l,i,n}(\omega, \mathbf{q}) = \frac{Q_0}{\sqrt{m_i}} \mathbf{e}_{i,n}(\mathbf{q}) e^{i(\mathbf{q} \cdot \mathbf{R}_l - \omega t)}, \quad (2)$$

where l denotes the unit cell, i is the atom's label in a unit cell, n is the branch of polarization, $\mathbf{q}$ is the wave vector, and ω is the angular frequency of the vibration. Q_0 is an arbitrary constant representing the arbitrary amplitude of normal mode. The position of the unit cell is denoted as $\mathbf{R}_l$ and t is the time. Atomic mass for the atom (l, i) is labeled as m_i . The eigenvector $\mathbf{e}_{i,n}$ is solved from the dynamic matrix $\|\mathbf{D} - \omega^2 \mathbf{I}\| = 0$ and satisfies the orthonormal relation $\sum_i \mathbf{e}_{i,n}(\mathbf{q}) \cdot \mathbf{e}_{i,n'}^*(\mathbf{q}) = \delta_{n,n'}$, where $\mathbf{D}$ is the dynamic matrix for each perfect lead, which is equivalent to the equation of motion Eq. (1) for the atoms in the unit cell on the lead. It is obvious that Eq. (2) automatically satisfies Eq. (1) for atoms in the lead. When we use the plane wave solution form Eq. (2), we do not need to consider the equation of motion Eq. (1) for atoms in the leads any more. That is the key to the scattering boundary method.

Then the vibrations on the lead can be seen as combinations of the refraction processes for normal-mode plane waves incident from each lead because of linearity of the system, which is similar to the case⁸ in continuum media. If a normal mode $\mathbf{u}_{l,i,n}^L(\omega, \mathbf{q})$ is incident from the left lead, the solution for the perfect leads can be assumed as

$$\mathbf{u}_{l,i}^{0,L} = \mathbf{u}_{l,i,n}^L(\omega, \mathbf{q}_n) + \sum_{n'} t_{n'n}^{LL} \mathbf{u}_{l,i,n'}^L(\omega, \mathbf{q}_{n'}'), \quad (3a)$$

$$\mathbf{u}_{l,i}^{0,R} = \sum_{n''} t_{n''n}^{RL} \mathbf{u}_{l,i,n''}^R(\omega, \mathbf{q}_{n''}'). \quad (3b)$$

Here the extra superscript L and R indicate the left and the right. In these equations, $t_{n'n}^{RL}$, $t_{n'n}^{LL}$ are the amplitude transmission and reflection coefficients from mode n on the lead L to mode n'' on lead R and to mode n' on lead L . The wave vectors $\mathbf{q}'$ and $\mathbf{q}''$ is found to satisfy $\omega = \omega_{n'}(\mathbf{q}') = \omega_{n''}(\mathbf{q}'')$. Note that the frequency does not change because the system is linear. The wave vectors are generally different for each mode in each lead. A similar expression can be written down for the right semi-infinite lead if a normal mode is incident from the right lead. Equation (3) is called the *scattering boundary condition*.

Dynamic matrix in the junction. The junction part usually does not have any translational symmetry. We cannot assume the plane wave solution for the vibrations of atoms in the junction part in contrast with that of atoms in the lead. We write the equation of motion, Eq. (1), for atoms in the junction part into a matrix form with the atom index i , which is similar to the $\tilde{k}$ in Ref. 6.

Scattering boundary equations. Combining Eq. (3) and the dynamic matrix in the junction, we write the dynamic matrix equations for atoms in the junction part and the scattering boundary conditions Eq. (3) into a matrix equation, illustrated as follows:

$$\begin{pmatrix} \cdots \\ \vdots & K_{ii} & -K_{ij} \\ 1 & & -u_{q',n}^L \\ & 1 & -u_{q'',n}^R \end{pmatrix} \begin{pmatrix} u_1 \\ \vdots \\ u_m \\ r_n \\ t_n \end{pmatrix} = \begin{pmatrix} 0 \\ \vdots \\ 0 \\ u_{q,n}^L \\ 0 \end{pmatrix}, \quad (4)$$

where $K_{ii} = -m_i \omega^2 + \sum_j K_{ij}$. Here, r_n and t_n denotes the reflected and transmitted amplitude transmission. The unknown amplitude transmissions can be solved from Eq. (4). We call the above method as the *scattering boundary equation method* (SBM). Note that Eq. (4) is actually equivalent to Eq. (1) and we just replace the equation of motion for atoms in the leads with the equivalent scattering boundary condition Eq. (3) with help of the periodical property of the leads.

For an epitaxial interface, only several atoms in a unit cell need to be connected directly through dynamic equations of motion due to the transverse translational symmetry. The present SBM is then equivalent to the previous approach in Refs. 4 and 5. Here, our extension of the scattering boundary equation method can cope with the *defect junction part* by the *incorporation of the dynamic matrix* for the atoms in the junction part.

Compared with the Green-function method in Ref. 6, the SBM method replaces the self-energy with the easily calculated eigenmode to compute energy transmission for each incident phonon mode, respectively. We think that the SBM method can be easily implemented and avoids the calculation of the surface Green's function, thus improving the computation efficiency. In addition, the energy transmission of one specific phonon mode to other different phonon modes can be clearly calculated and presented by the SBM.

The obvious difficulty for solving Eq. (4) is that it may not be a square matrix equation. However, the boundary conditions given for atoms on leads are not independent. Some equations from the boundary conditions are redundant. They are, nevertheless, consistent equations and still have a unique solution. This consistency can be verified through the method of Gaussian elimination for Eq. (4). The standard singular value decomposition (SVD) method⁹ can identify the independent equations and solve them efficiently.

B. Energy transmission expression

Once the amplitude transmissions $t_{n'n}^{LL}, t_{n''n}^{RL}$ have been solved through the SBM from Eq. (4), we need to find the energy transmission expression from these amplitude transmissions. As we have mentioned in the Introduction, some energy transmission expressions have been proposed for the lattices with identical atomic mass on each lead and equal lattice constant. Here, we present a more general energy transmission expression, starting from the energy flux formula for lattice.

A time-average energy flux $\bar{I}_z$ along the z direction for the perfect lead can be easily written as

$$\bar{I}_z = \frac{-i}{V} \sum_{\mathbf{q},n} \omega_n(\mathbf{q}) \frac{\partial \omega_n(\mathbf{q})}{\partial q_z} \langle \dot{Q}_q^n \dot{Q}_q^{n*} \rangle_t, \quad (5)$$

where V is the lead volume and Q_q^n is the normal-mode coordinates for the n th polarized normal-mode wave with momentum $\mathbf{q}$. In the above expression, $\dot{Q}_q^n$ is the normal-mode coordinates differentiated with respect to time and the bracket $\langle \rangle_t$ means the time average. Since energy must be conserved and there is no net energy accumulation in the junction, the time-averaged energy currents of both sides must be equal,

$$\mathbf{I}^L \equiv \mathbf{I}^R, \quad (6)$$

where $\mathbf{I}^\sigma = \bar{I}^\sigma \cdot S_\sigma$, $\sigma = L, R$, and S_σ represents the cross section area of each lead. Note that it is the energy currents $\mathbf{I}^\sigma$ that should be equal for the two sides. The conservation of the current yields the following relation for the transmission amplitudes by following a similar approach for the continuum case:⁸

$$\sum_{\sigma,n} t_{nn'}^{\sigma\sigma'} t_{nn''}^{*\sigma\sigma''} \bar{v}_n^\sigma = \bar{v}_{n'}^{\sigma'} \delta_{n'n''} \delta_{\sigma'\sigma''}, \quad \bar{v}_n^\sigma = v_n^\sigma / l_z^\sigma, \quad (7)$$

where l_z^σ is the length of unit cell along the z direction, $v_n^\sigma = \partial \omega_n^\sigma / \partial q_z$. This equation is analogous to the unitary condition for the scattering matrix for electronic transmission.¹⁰ Equation (7) is also similar to that of continuum case,⁸ except the difference that *group velocity* should be replaced by a *reduced group velocity* $\bar{v}_n^\sigma = v_n^\sigma / l_z^\sigma$. This is a quite natural result for lattice dynamics. When we consider the details of lattice wave, the lattice constant comes to view.

From Eq. (7), the energy transmission, for example, from mode (L, n) to mode (R, n'') , is given by

$$\tilde{T}_{nn''}^{RL} = |t_{nn''}^{RL}|^2 \frac{\bar{v}_{n''}^R}{\bar{v}_n^L}. \quad (8)$$

The total reflection and transmission coefficients for (L, n) are given, respectively, by

$$\mathcal{R}_n^L = \sum_{n'} |t_{nn'}^{LL}|^2 \frac{\bar{v}_{n'}^L}{\bar{v}_n^L}, \quad \mathcal{T}_n^L = \sum_{n''} |t_{nn''}^{RL}|^2 \frac{\bar{v}_{n''}^R}{\bar{v}_n^L}, \quad (9a)$$

$$\mathcal{R}_n^L + \mathcal{T}_n^L \equiv 1. \quad (9b)$$

The above energy conservation relation, Eq. (9b), can be automatically satisfied if the algorithm for calculating the amplitude transmission is correct. Note that we should choose the *same* constant Q_0 in Eq. (2) for *all* incident, reflected, and transmitted lattice waves when using Eq. (9b).

We can clarify the relation of our formula Eq. (9a) for energy transmission with $t_{\mathbf{k}j} = \rho_A \sum_q v_{qz} A_q^2 / (\rho_B v_{0z} A_0^2)$ in Ref. 4 and $\mathcal{T} = M_2 \sum_s |t_s|^2 v_s^{(i)} / M_1 |v^{(i)}|$ in Ref. 5. Here $t_{\mathbf{k}j}$ and $\mathcal{T}$ are the energy transmissions across the solid interface and they have the same meaning as our notation T_n^σ . ρ_B and ρ_A are mass densities for the leads of incoming and outgoing waves, while M_1 and M_2 are atomic masses. A_0 is the amplitude for the incoming wave and A_q is the amplitude for the transmitted wave, where q denotes the polarization branch. t_s is the

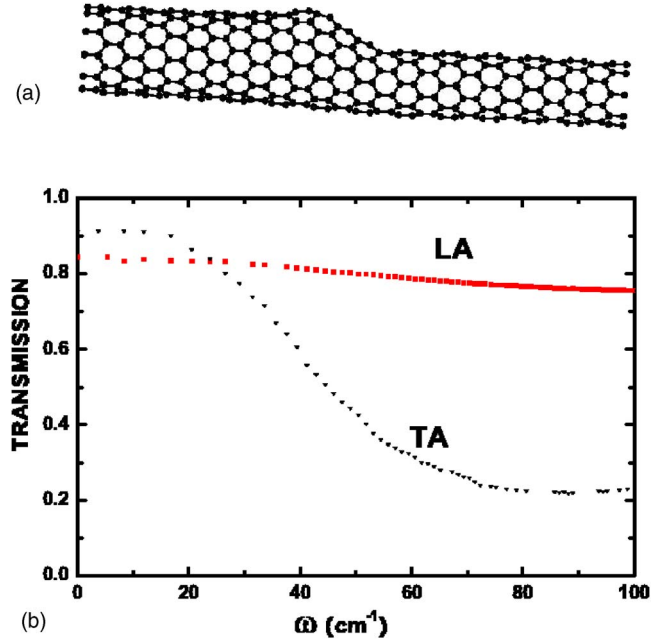


FIG. 1. (Color online) (a) Structure of an (11,0) and (8,0) nanotube junction. (b) The energy transmission $T_n(\omega)$ as a function of angular frequency ω .

amplitude for transmitted waves. Reference 5 has set the incoming amplitude to 1. v_{qz} and $v_s^{(i)}$ are the group velocities of transmitted waves along the direction normal to the interface, while v_{0z} and $v^{(i)}$ are the group velocities of incident waves along the same direction. Provided that atomic masses in unit cell are identical, they can be factored out. Then, we find that the relation of the amplitude transmission A used in Ref. 4 with our Eq. (9a) is just that $A_{\sigma \rightarrow \sigma'} \rightarrow t^{\sigma\sigma'} / \sqrt{m_\sigma}$, where $\sigma = L, R$. With this relation, $t_{\mathbf{k}j}$ and T_n^σ are consistent with Eq. (9a), respectively, for the models they studied. They are valid in the case for unit cells with *identical* masses. Here, we have extended the energy transmission calculation for composite lattices, where atomic masses in unit cell for most materials need not be equal. Also the appearance of the lattice constant in our formula is quite natural because the lattice wave can feel the difference in lattice spacing.

III. MODEL AND NUMERICAL RESULTS

Next, Using the SBM of Eq. (4) and the energy transmission formula Eq. (9), we compute the energy transmission across the nanotube junctions. The semiconductor nanotube junction structure (11,0)–(8,0) is first constructed by a geometrical method as in Ref. 2. However, there are many defects in the geometrically constructed structure. We optimize the junction structure by a second-generation Brenner potential¹¹ to relax the atoms to their equilibrium positions. The optimized structure is shown in Fig. 1(a). The force constants are computed numerically by a finite difference method from the same potential. The phonon dispersion for nanotube (11,0) calculated from these linearized force constants is illustrated in Fig. 2. Four acoustic branches are considered for energy transport: the longitudinal mode (LA),

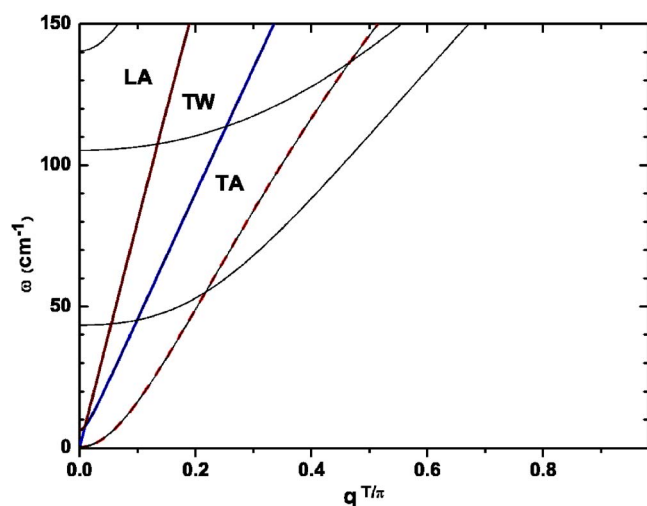


FIG. 2. (Color online) Phonon dispersion for nanotube (11,0). The wave number q is expressed in terms of the length of nanotube translation vector T .

doubly degenerate transverse mode (TA), and the unique twist mode (TW) in nanotubes.

Following the SBM in the previous section, the SBM equations are constructed with 504 equations and 455 variables. The boundary conditions given for atoms on leads are not independent. Some equations from the boundary conditions are redundant because the motion for each boundary atom is correlated together for a specific polarized mode. The number of variables is equal to the independent equations in the above SBM equation set. We have numerically verified the consistency of these equations by calculating the rank of SBM equations with the Gauss elimination for this equation set. The rank we obtained is 455 within numerical accuracy, which matches the number of variables. The SBM equations are *consistent* equations and have a *unique* solution. These equations are then solved through the standard singular value decomposition method⁹ as mentioned.

The transmission coefficients for the nanotube junction are shown in Fig. 1(b). We did not find mode-mixing or mode-conversion behavior among acoustic modes at the low-frequency range although all modes were considered. For example, the reflected and the transmitted waves across the junction for the incident LA mode are only LA modes. This is due to the high symmetry of atomic motion for nanotubes. Each traveling wave on nanotubes has its own symmetrical property resulting from the symmetry of the nanotube structure. The symmetries on the left and right leads are different. It is hard to convert one particular symmetrical motion to other symmetrical motions across the conjunction unless the symmetries on each lead are the same. This kind of mismatch in the symmetry of motion of atoms on nanotubes will play an important role in the thermal conduction of nanotube mixtures.

The LA modes are the common symmetrical motions for both the left and right leads. The transmission for the LA mode stays around 0.8 with only some small changes. This value is below the acoustic-mismatch model (AM) prediction¹² of 0.98 with the longitudinal group velocity 20.18

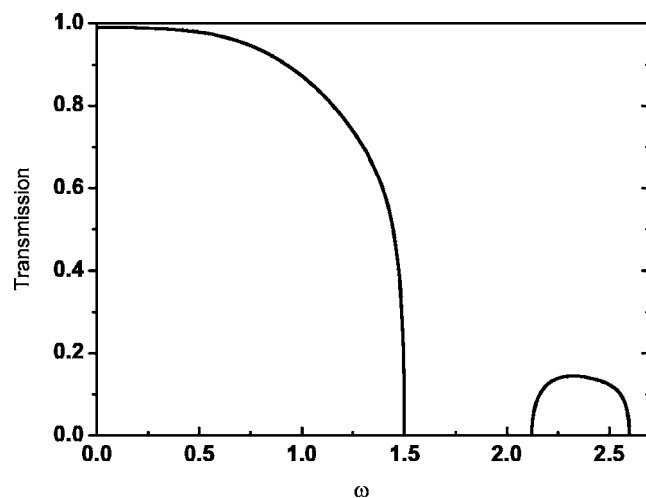


FIG. 3. Energy transmission on an acoustically mismatched 1D chain.

and 20.95 km s⁻¹ for (11,0) and (8,0). In contrast, the transmissions of the TW mode and many other optical modes are nearly zero or very small. The transmission for the TA mode decreases with frequency. Two reasons can account for this: (i) Although the TA mode is a common symmetry for both left and right leads, the kind of transverse symmetry is destroyed by the central junction part. It can be seen from Fig. 1 that the central junction part does not have transverse symmetry. (ii) There is a mismatch of dispersion relation for TA mode because the TA mode has a nearly quadratic dispersion relation, as illustrated in Fig. 2. These factors will inhibit the transmission for TA mode at higher frequency.

To estimate the maximum ratio of the possible acoustic-optical mode conversion, we consider a simplified 1D toy chain model because it can be assumed that the acoustic-optical conversion will be easier for both longitudinal waves. The unit cell in the left lead comprises two atoms with different masses m_1 and m_2 . The right side chain comprises a simple lattice with atomic mass m_3 . The two chains are connected through two atoms in the central part with masses m_{f1} and m_{f2} . The force constants are assumed to be K for all the springs. The lattice constants are a for the left chain and b for the right chain, respectively. There are two branches of normal modes for the left diatomic lead. We take $m_1=4$, $m_2=8$, $m_3=4$, $m_{f1}=3$, $m_{f2}=10$, $a=8$, $b=4$, and $K=9$, we obtain the energy transmission on this lattice chain shown in Fig. 3. It is interesting to observe that there is a mode conversion between a longitudinal acoustic mode and a longitudinal optical mode in the frequency range from about 2.12 to 2.60. But the optical-acoustic transmission is much smaller than the acoustic-acoustic transmission, no more than 20%. It is reasonable to conclude that the mode conversion between different polarized phonon modes may be trivial, especially for the highly symmetrized phonon modes in nanotubes.

It can be seen from the above that energy transmission across nanotube junctions is strongly dependent on the symmetrical property of the lead nanotube. Such mismatch in symmetries will restrict the number of possible transmission modes and thus blocks the thermal conduction across the junction. We also suggest that this kind of mode-dependent

transmission behavior may be important for further application such as phonon filters. It may also be interesting for the improvement of nanotube thermal conduction by amending the interface between nanotube and its substrate.

IV. CONCLUSION

In summary, a scattering boundary equation method (SBM) is proposed for the calculation of the amplitude transmission across solid junctions. A general energy transmission expression for lattice waves across solid junctions is also given. Its relation with atomic masses, lattice constants, and group velocities is elucidated. Our calculation shows a strongly mode-dependent transmission in nanotube junction due to the symmetrically vibrating motion for nanotube atoms, indicating its possible important role in nanotube thermal conduction. In addition, we remark that (i) The SBM

needs translational periodicity in the lead or that the lead should be perfect crystal. The SBM method fails if there is no traveling plane wave solution for the nonperiodical leads. In addition, the SBM method also assumes that the junction is small in comparison with the leads so that the lead can be modeled as semi-infinite. (ii) The lattice dynamic approach to solid junction is only within the linear approximation that may be accurate at low temperatures. However, the nonlinear effect or anharmonic effect has to be considered for high temperatures. To include such inelastic scattering will be an interesting problem to be pursued,¹³ but it cannot be covered in the present approach.

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