Eikonal approximation in atom-surface scattering: Effects of a corrugated attractive well

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The eikonal approximation, which is an extremely useful method of calculating intensities for the scattering of atomic beams from surfaces, is extended to include a periodic corrugation of the leading edge of an attractive square-well potential placed in front of the hard repulsive wall. This provides a method for estimating small effects of corrugation of the attractive physisorption potential on the diffraction spectra. Calculations indicate that the relative phase of the attractive well corrugations, with respect to those of the hard repulsive wall, has a distinctive and characteristic effect on the diffraction intensities.

I. INTRODUCTION

The eikonal approximation is one of the earliest semiclassical approaches that was applied to quantum-mechanical problems, and has been useful ever since for scattering calculations.[1] The eikonal approximation has been especially useful in atomic scattering from surfaces since its initial introduction to that field by Garibaldi *et al.*[2] for the case of scattering from a hard corrugated repulsive wall. It was immediately recognized that, in spite of its severe approximative nature, the combination of the eikonal approximation with a hard repulsive wall was capable of producing useful qualitative predictions of experimental diffraction spectra with very little calculation effort. When extended to include a square attractive well in front of the repulsive wall to mimic the effects of the physisorption potential, this model was shown early on to be capable of giving reasonable quantitative agreement with experimentally measured diffraction peak intensities.[3]

The eikonal approximation has been applied to both elastic[4–7] and inelastic scattering of atoms from surfaces,[8] and it can be adapted to describe scattering from either periodic or nonperiodic surfaces.[9–12]

Since the initial introduction of the eikonal approximation other theoretical methods have been developed to produce numerically exact solutions to the problem of atom diffraction from a corrugated repulsive hard wall.[13] There also now exist several methods for obtaining numerically exact solutions to the problem of diffractive scattering from a completely realistic periodic surface potential, including the coupled-channels method,[14] summing the perturbation series to high orders,[15] and wave-packet propagation.[16] Nevertheless, despite its seemingly apparent drawbacks due to its severe approximative nature, the eikonal approximation remains a useful tool because of its calculational ease and its remarkable ability to predict qualitative responses to changes in experimental parameters.[7,12,17,18]

In this paper we wish to address the question of how a corrugation appearing in the attractive physisorption well will affect the overall diffraction pattern in atom-surface scattering. The question of possible corrugations of the well and their effect on the diffraction pattern has a long history,[19] and it has been argued that the effects could be particularly strong in the case of bound-state resonances (selective absorption).[20]

Examples of crystal surfaces where the adsorption well might be strongly corrugated are molecular crystals containing strongly polarizable atoms. A case in particular is the (001) surface of MgO, which is strongly corrugated but the corrugation is dominated by the oxygen ions and not by the smaller Mg ions. Since the repulsive force on an incoming atomic scattering projectile is due to Pauli exclusion of the overlapping electronic distributions, the repulsive corrugation will be due mostly to the oxygen atom. However, in the well one would also expect a strong corrugation of the attractive potential due to the large polarizability of the oxygen ions. In fact, one could expect that the polarization-induced corrugation in the well might be out of phase with the repulsive corrugation, i.e., the well might have a deeper minimum directly in front of an oxygen ion at the same lateral position where, closer to the surface, the repulsive corrugation would have its maximum.

The particular question addressed here is to show how the eikonal method can be extended to include a corrugation in the attractive potential well in front of the repulsive hard wall. In particular, we show how the eikonal method can be readily extended to include a corrugation of the leading edge of a square-well potential. Although a square well is a rather crude approximation to the correct form of the adsorption potential, which has a long-range attractive part behaving as $1/z^3$ where z is the perpendicular distance from the surface, it is expected that such a model will provide useful estimates of the characteristic behavior to be expected from changes in the physical parameters of the well.

The approach taken here is based on methods used in classical diffraction of sound or electromagnetic waves, in either transmission or reflection mode, from a two-dimensional optical ''phase grating.'' This phase grating approach provides an alternative method for deriving the well-known theoretical expressions for the eikonal approximation.

The paper is organized as follows. In the next section the phase grating approach is discussed and a derivation of the

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eikonal approximation is developed in that context. In Sec. III the eikonal formalism is extended to include a general periodic corrugation of the leading edge of the well. In Sec. IV results are presented for the special case of sinusoidal corrugations where all expressions can be evaluated analytically. In Sec. V we carry out example calculations and in Sec. VI discuss some conclusions.

II. THE EIKONAL APPROXIMATION

For simplicity, we will consider a hard repulsive wall with a one-dimensional corrugation; the z coordinate is perpendicular and the x coordinate parallel to the surface. The extension to higher-dimensional corrugations is trivial. The corrugation of the surface is defined by $z=\xi(x)$ where $\xi(x)$ is the corrugation function, and the condition of periodicity is $\xi(x+na)=\xi(x)$ where n is an arbitrary integer and a is the corrugation period. Thus the interaction potential is defined by

$$V(x,z)=\begin{cases}\infty, & z\leqslant\xi(x)\\ 0, & z<\xi(x)\end{cases}. \tag{1}$$

The asymptotic form of the Schrödinger wave function for a scattered particle with incident plane-wave boundary conditions must be in the form of a Bloch function and can be written as

$$\Psi(x,z)\to e^{iK_ix-ik_{iz}z}+\sum_G C(G)e^{i(K_i+G)x+ik_{Gz}z}, \tag{2}$$

where K_i and k_{iz} are the x and z components of the incident momentum, respectively, and the translational energy is $E_i=\hbar^2(K_i^2+k_{iz}^2)/2m$, where m is the projectile mass. The reciprocal lattice vectors G are given by $G=2\pi n/a$ where $n=0,\pm1,\pm2,\ldots$. The final perpendicular wave-vector components k_{Gz} are determined by conservation of energy and parallel momentum and are given by $k_{Gz}=\sqrt{K_i^2+k_{iz}^2-(K_i+G)^2}$, where the positive value of the square root is taken. Because of the extreme short-range nature of the hard corrugated wall potential, the asymptotic form of Eq. (2) is valid for all z outside the selvedge region, i.e., for $z>\mathrm{Max}|\xi(x)|$.

The traditional manner of developing the eikonal approximation is to first apply the Rayleigh ansatz,[2,21] which is to assume that the asymptotic solution of Eq. (2) can be extended into the selvedge region right up to the hard repulsive wall. Then application of the boundary condition

$$\Psi(x,z=\xi(x))=0 \tag{3}$$

and making the eikonal assumption that k_{Gz} varies slowly with G leads immediately to a simple evaluation of the diffraction amplitude given by

$$C(G)=\frac{-1}{a}\int_0^a dx\, e^{-iGx}e^{-i(K_{iz}+K_{Gz})\xi(x)}. \tag{4}$$

The overall factor of -1 on the right-hand side of Eq. (4) is the phase factor $\exp(i\pi)$ expected from a hard-mirror collision, and $k_{iz}+k_{Gz}$ is the normal momentum transfer in the collision. Evanescent diffraction beams are ignored in the eikonal approximation.

An alternative approach to the eikonal approximation, which leads to the same result as Eq. (4), is to regard the surface as a classic Fraunhofer diffraction problem of scattering from a phase grating. This will be the approach developed for the attractive square well in Sec. III below. In the scattering of a scalar wave field from a one-dimensional periodic diffraction grating of period a, the asymptotic form of either the transmitted or reflected wave is given by[22]

$$\Psi(x,z)\to\sum_G A(G)e^{i(K_i+G)x\pm ik_{Gz}z}, \tag{5}$$

where the diffraction amplitude is given by

$$A(G)=\frac{1}{a}\int_0^a dx\, e^{-iGx}t(x), \tag{6}$$

and $t(x)$ is the transmission function. As an example, for a periodic transmission grating in the primitive Kirchhoff approximation $t(x)=t(x+na)$ with $n=0,\pm1,\pm2,\ldots$, and $t(x)$ takes only two values, $t(x)=0$ at the positions of the opaque grating bars and $t(x)=1$ at the positions of the transparent slits. Alternatively, one can write $t(x)=\exp[i\phi(x)]$, where $\phi(x)$ is the phase gained by the wave at each point x along the grating. This alternative approach is called a phase grating, and the amplitude of Eq. (6) becomes

$$A(G)=\frac{1}{a}\int_0^a dx\, e^{-iGx}e^{i\phi(x)}. \tag{7}$$

To apply this to the eikonal problem, we need to determine the appropriate phase function. In order to do this, one picks a point (x_1,z_1) above the surface and allows the incoming wave to propagate toward the surface, collide with the surface, and then propagate back to that same point. The total phase gained by a plane wave in such a process is

$$\phi(x)=(k_{iz}+k_{Gz})z_1-(k_{iz}+k_{Gz})\xi(x)+\pi. \tag{8}$$

The final term π on the right-hand side of Eq. (8) arises from the reflection from a hard-mirror surface, and the first term is a trivial constant. Thus when Eq. (8) for the phase factor is inserted back into the amplitude (7), apart from a trivial phase the result is identical with the standard eikonal result of Eq. (4). It is this ''phase grating'' approach that is used below to develop the eikonal approximation for a corrugated attractive well in front of the surface.

III. CORRUGATED ATTRACTIVE WELL

We now wish to extend the eikonal approximation of Sec. II to include an attractive well of uniform depth D in front of the corrugated hard wall, and to allow for the leading edge of this attractive well to also be corrugated with a corrugation function $\eta(x)$. Such a potential is defined by

$$V(x,z)=\begin{cases}0, & z>b+\eta(x)\\ -D, & b+\eta(x)\geqslant z>\xi(x)\\ \infty, & z\leqslant\xi(x),\end{cases} \tag{9}$$

where b is the width of the attractive well. If both corrugation functions $\eta(x)$ and $\xi(x)$ are periodic with period a then the asymptotic form of the wave function for $z>b+\mathrm{Max}|\eta(x)|$ is the same as Eq. (2). The wave function in the region inside the well will also be of Bloch form, but will consist of diffraction beams traveling in both the positive and negative z directions,

$$\Psi(x,z)=\sum_G F(G)e^{i(K_i+G)x-i\tilde{k}_{Gz}z} + \sum_G H(G)e^{i(K_i+G)x+i\tilde{k}_{Gz}z}, \tag{10}$$

where $\tilde{k}_{Gz}=\sqrt{K_i^2+k_{iz}^2-(K_i+G)^2+2mD/\hbar^2}$ is the perpendicular wave vector inside the well. In the eikonal approximation, backward scattering of the wave by the leading edge of the attractive square well is ignored. The transmission amplitude coefficient $F(G)$ is then calculated using the phase grating approach developed in Sec. II above. The phase change for transmission across the leading edge of the corrugated well is $\phi_W(x)=(\tilde{k}_{Gz}-k_{iz})\eta(x)$, which gives from Eq. (7)

$$F(G)=\frac{1}{a}\int_0^a dx\, e^{-iGx}e^{i(\tilde{k}_{Gz}-k_{iz})\eta(x)}. \tag{11}$$

The incoming wave in the attractive well (10) now consists of all possible real diffraction beams, and each of these diffraction beams upon collision with the corrugated hard wall acts as the source of a new series of backscattered diffraction beams. Summing all of these outgoing diffraction beams gives the outgoing amplitude in the well as

$$H(G)=\sum_{G'} e^{i\tilde{k}_{G'z}b}F(G')E(G,G'), \tag{12}$$

where the scattering amplitude generated by each of the incoming diffraction beams is determined from the phase grating expression of Eq. (7) to be

$$E(G,G')=\frac{-1}{a}\int_0^a dx\, e^{-i(G-G')x}e^{-i(\tilde{k}_{Gz}+\tilde{k}_{G'z})\xi(x)}. \tag{13}$$

The final operation is to allow the outgoing diffraction beams in the well to traverse the leading edge where, once again, each diffraction beam acts as the source of a complete series of outgoing beams. As before, consistent with the eikonal approximation, only transmission across the leading edge is considered and reflection of waves back into the well is ignored. Again applying the phase grating method (7), the final outgoing scattering amplitude of the asymptotic wave function (2) is found to be

$$C(G)=\sum_{G'} e^{i\tilde{k}_{G'z}b}H(G')D(G,G'), \tag{14}$$

where

$$D(G,G')=\frac{1}{a}\int_0^a dx\, e^{-i(G-G')x}e^{-i(\tilde{k}_{G'z}-k_{Gz})\eta(x)}. \tag{15}$$

The final scattered intensities in each diffraction beam $I(G)$ are given by the usual expression

$$I(G)=\frac{k_{Gz}}{k_{iz}}|C(G)|^2, \tag{16}$$

and the unitarity sum is

$$U=\sum_G I(G), \tag{17}$$

where the summation over reciprocal lattice vectors is limited to real open diffraction beams. For an exact theory, $U=1$, but for approximate theories this will not necessarily hold. Nevertheless, when the eikonal theory is a reasonable approximation, U will be a number close to 1, and this serves as a useful check on the validity of the approximation.[2]

IV. SINUSOIDAL CORRUGATION FUNCTIONS

A particularly useful expression of historical importance in problems involving hard corrugated potentials is the sinusoidal function.[2,21,23] In this case the corrugation function of the hard repulsive wall is given by

$$\xi=h_R a\cos\left(\frac{2\pi}{a}x\right), \tag{18}$$

where h_R is the dimensionless corrugation amplitude of the hard wall in units of the period a, and that of the leading edge of the well is given by

$$\eta=h_W a\cos\left(\frac{2\pi}{a}x\right), \tag{19}$$

where h_W is the corrugation amplitude of the well. This form of the corrugation allows exploitation of the integral representation for the Bessel function,

$$J_n(y)=\frac{i^n}{2\pi}\int_0^{2\pi} d\theta\,\exp[-i(y\cos\theta\pm n\theta)], \tag{20}$$

where n is a positive integer and $J_{-n}(y)=J_n(-y)=(-1)^n J_n(y)$.

For the corrugation functions of Eq. (19) the scattering amplitude $F(G)$ of Eq. (11) becomes

$$F(G)=i^g J_g([\tilde{k}_{Gz}-k_{iz}]h_W a), \tag{21}$$

where the integer g is related to the reciprocal lattice vectors by $G=2\pi g/a$, and if either $g<0$ or $\tilde{k}_{Gz}-k_{iz}<0$ the right-hand side of Eq. (21) is to be multiplied by $(-1)^g$.

Similarly, the amplitude $E(G,G')$ of Eq. (13) is given by

$$E(G,G')=-i^{g'-g}J_{g-g'}([\tilde{k}_{Gz}+\tilde{k}_{G'z}]h_R a), \tag{22}$$

and if $g-g'<0$ then the right-hand side of Eq. (22) is to be multiplied by $(-1)^{g-g'}$.

Finally, the amplitude $D(G,G')$ of Eq. (15) is given by

$$D(G,G')=i^{g'-g}J_{g-g'}([\tilde{k}_{G'z}-k_{Gz}]h_W a), \tag{23}$$

where once again the right-hand side of Eq. (22) is to be multiplied by $(-1)^{g-g'}$ if $g-g'<0$ or if $\tilde{k}_{G'z}-k_{Gz}<0$.

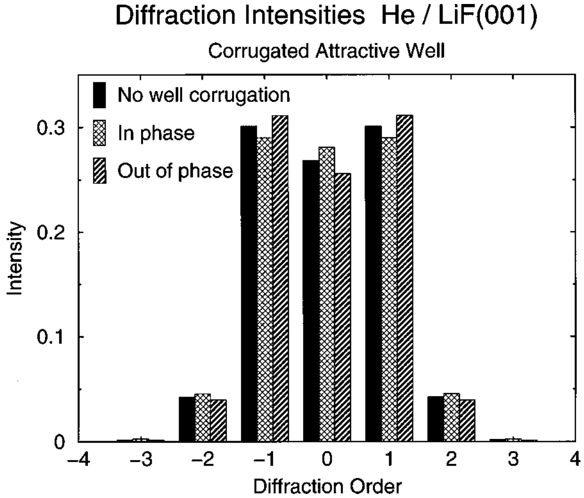

FIG. 1. Diffraction intensities for a one-dimensional sinusoidal corrugation with parameters corresponding to He atom scattering from LiF(001). The beam is normally incident and the energy is $E_i = 63$ meV. The solid vertical bars are calculations for no corrugation of the well, the cross-hatched bars are for the well corrugation the same as that of the hard wall and in phase, and the hatched bars are for the same well corrugation amplitude but out of phase.

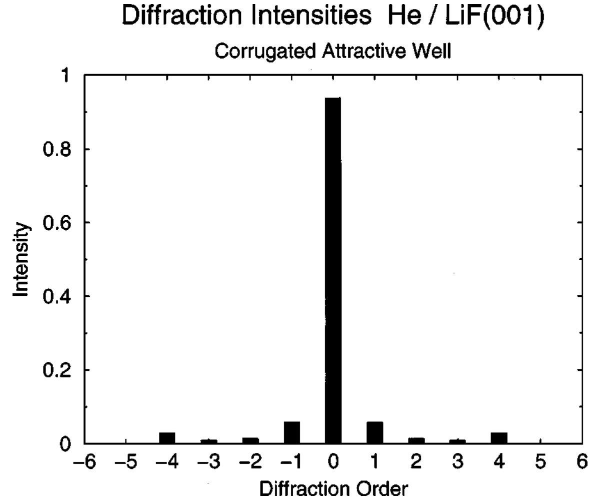

FIG. 2. Diffraction intensities for a one-dimensional sinusoidal corrugated well. The beam is normally incident and the energy is $E_i = 63$ meV as in Fig. 1. The repulsive hard wall is flat and uncorrugated and the leading edge of the well has a large corrugation amplitude $h_W = 0.23$.

Thus in the case of sinusoidal corrugations Eqs. (21)–(23) permit the solution to be written in compact form in terms of simple Bessel functions.

V. CALCULATIONS

For carrying out example calculations, a very useful model is the application of the eikonal approximation to the scattering of He and Ne from the (001) face of LiF carried out by Boato, Cantini, and Mattera.[3] This calculation was used to determine the corrugation function of the LiF surface for the two different atomic projectiles. In the case of He or Ne atom scattering from cleaved LiF(001) the corrugation of the repulsive potential is caused nearly entirely by the large F^- ions with negligible effect of the much smaller Li^+ ions. The corrugation appears as a square two-dimensional sinusoidal corrugation with a period $a = 2.84$ Å. For He scattering with an incident energy $E_i = 63$ meV, Boato *et al.* found a well depth of $D = 5$ meV and a one-dimensional cut across the surface in the direction of a close-packed row had a corrugation amplitude $h_R = 0.023$.

Calculations using the parameters of Boato *et al.* for a one-dimensional corrugation are shown in Fig. 1 for He atom scattering with incident energy $E_i = 63$ meV, perpendicular incidence, and a well width $b = 3$ Å. The solid vertical bars in Fig. 1 show the diffraction peak intensities as a function of diffraction order for an uncorrugated well. The cross-hatched vertical bars are diffraction intensities for a well corrugation equal to that of the hard corrugated wall, $h_W = h_R = 0.023$, and both corrugations are in phase. The hatched vertical bars are intensities calculated with the same well corrugation amplitude but of opposite phase from that of the hard wall, i.e., $h_W = -h_R = -0.023$. The unitarity summation in all of these calculations was $U = 0.96$. This is a rather large corrugation and the diffraction is quite strong, with the ± 1 order diffraction peak intensities larger than that of the specular. The effect of the well corrugation is seen to be a small perturbation on the intensities of $\approx 10\%$ or less. However, on each of the peaks there is a very distinct difference in the effect of an ''in-phase'' and an ''out-of-phase'' well corrugation. If the ''in-phase'' well corrugation increases the intensity of a particular peak, then reversing the phase of the well corrugation decreases the intensity of that same peak, and vice versa. This effect is clearly observed in the specular as well as the first- and second-order peaks in Fig. 1. Numerous calculations with varying incident energies, different angles of incidence, and different corrugation amplitudes have confirmed that this is a general characteristic effect.

Figure 2 is a calculation similar to Fig. 1, also at normal incidence and with $E_i = 63$ meV but with no corrugation of the hard repulsive wall, $h_R = 0$. The corrugation of the well has been increased by an order of magnitude, $h_W = 0.23$, in order to show the effect of the well corrugation alone. The ± 1 order peaks have intensities of approximately 5% of the specular intensity, and all other diffraction peaks are nearly negligible. By contrast, a very similar diffraction pattern is obtained with the well corrugation set to zero and a hard-wall corrugation of only $h_R = 0.008$. This comparison indicates that the corrugation of the repulsive hard wall is roughly 30 times as effective in creating intensity in the diffraction peaks as a corrugation in the attractive well. This effect can be clearly understood from a comparison of the scattering amplitude of Eq. (11), which describes traversal of the wave across the leading edge of the well, and that of Eq. (13), which describes the backward reflection from the repulsive wall. In the case of the reflection from the repulsive wall the total perpendicular momentum transfer is the sum of the initial and final perpendicular momenta and this gives a very large phase in the integrand of Eq. (13). However, in the case of traversal of the well, the perpendicular momentum transfer is the difference of the final and initial perpendicular momenta and hence leads to a much more slowly varying phase in the integrands of Eqs. (11) and (15).

It is of interest to explore the limits of validity of the eikonal approximation with respect to the size of the corrugation amplitude. This can be tested by increasing the corrugation amplitude until the unitarity begins to differ substantially from the value 1. Setting $h_W = h_R = 0.06$ gives a unitarity value of 0.8. For larger corrugation parameters the unitarity value rapidly becomes substantially worse than 1, so this appears to be a crude upper limit on the validity of these calculations.

VI. CONCLUSIONS

In this paper the eikonal approximation as applied to elastic atom-surface scattering has been reformulated in terms of the theory of scattering by a phase grating as commonly applied in sound wave or optical wave scattering. This formulation of the eikonal approximation has been used to solve for the diffraction intensities generated by a monoenergetic incident beam of atoms scattering from a hard corrugated wall having an attractive square adsorption well with a corrugated leading edge. This solution is used as a model for estimating the effects of corrugation within the attractive adsorption well and to compare effects of the well corrugation with those of the corrugation of the repulsive part of the potential.

Such a corrugated square-well model is expected to overestimate the effects on the intensity of a more realistic corrugated well potential with the correct $1/z^3$ behavior of the long-range Van der Waals attraction. However, because of the simplicity of this formalism and the ease of calculations it is expected that this solution will be useful for predicting physical trends, just as the ordinary eikonal approximation is still very useful for obtaining crude theoretical estimates. An even simpler formalism, expressed entirely in terms of Bessel functions, results in the case of purely sinusoidal corrugations for the repulsive wall and leading edge of the well.

Several example calculations were carried out, which demonstrate that the corrugation of the leading edge of the square well has an effect on the diffraction intensities that is about 5% as strong as that of an equally large corrugation of the repulsive wall. An interesting question that can be answered with this formulation concerns the effect of a well corrugation that is in or out of phase with the corrugation of the repulsive wall. The present calculations show that there is a very characteristic signature of the relative phase of the well corrugation with respect to the corrugation of the repulsive wall. If, when compared to a calculation with an uncorrugated well, the addition of corrugation to the well increases (or decreases) the intensity of a particular diffraction peak, then changing the phase of the well corrugation by 180° will reduce (or increase) the intensity of that same peak.

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