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Short Communications

Atom–Surface Elastic Scattering in the Presence of Laser Radiation*

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A qualitative discussion on atom–surface elastic scattering taking place in the presence of laser radiation is given. It is suggested that appreciable effects of laser radiation on diffraction patterns may be expected if the laser radiation is capable of inducing electronic transitions in atoms with a large probability.

Key words: Scattering, atom–surface elastic ~ – Laser radiation

We wish to point out that atoms in different electronic states may produce vastly different diffraction patterns when scattered off solid surfaces. This immediately suggests that, when scattering takes place in the presence of laser radiation which is capable of inducing electronic transitions in atoms, we may expect appreciable effects of laser radiation on diffraction patterns.

Let us first consider scattering of electronically excited atoms from solid surfaces in the absence of laser radiation. At the present time, accurate quantitative treatments of this problem are not possible because of the lack of information on excited atom–surface potential curves. However, a semi-quantitative treatment can be given with the help of an approximate method recently developed by us [1] to describe high-energy atom–surface diffractive scattering. Let us assume that atoms prepared in

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their ith electronic state are normally incident on a periodic surface with sufficiently high energy so that our method is valid. The natural lifetime of the ith state is assumed to be long compared with the scattering time. We also assume that the potential energy between the atom in the ith state and the surface is given by the Lennard–Jones–Devonshire potential,

$$V_i(XYZ) = V_{0_i}(Z) + V_{1_k}(Z) \cos\left(\frac{2\pi X}{a} + \cos\frac{2\pi Y}{a}\right), \tag{1a}$$

$$V_{0_i}(Z) = D_i \exp\left[-\alpha_i(Z - Z_0)\right]\{\exp\left[-\alpha_i(Z - Z_0)\right] - 2\}, \tag{1b}$$

$$V_{1_i}(Z) = -2\beta_i D_i \exp\left[-2\alpha_i(Z - Z_0)\right], \tag{1c}$$

where a is the lattice spacing, and the subscript i in the potential parameters D, α and β refers to the ith electronic state. The Lennard–Jones–Devonshire potential does not accurately represent the actual potential for highly excited atoms for which the potential is repulsive throughout. Although this case can be treated with only a slight modification, we restrict our attention to low-lying excited states for which the Lennard–Jones–Devonshire potential represents a good approximation. The normalized intensity of the (mn)th diffraction peak is then given by [1]

$$P_{mn} = J_m^2[u_i(E)]J_n^2[u_i(E)], \tag{2}$$

where

$$u_i(E) = -\frac{4\beta_i E}{\hbar\gamma_i}\left\{1 + \sqrt{D_i/E}\left[\frac{\pi}{2} + \sin^{-1}\left(\sqrt{D_i/(D_i + E)}\right)\right]\right\}, \tag{3}$$

$$\gamma_i = \frac{2\alpha_i^2 E}{\mu}; \tag{4}$$

E is the incident energy, and μ is the mass of the atom. The notation $u_i(E)$ signifies that the argument of the Bessel functions depends on E as well as on the initial preparation of the atom in the ith electronic state. Since P_{nm} as given by Eq. (2) strongly depends on the argument of the Bessel functions, it may be concluded that a different incident energy or a different electronic state (with which different values of D, α and β are associated and consequently a different value of the argument is associated) may produce significantly different diffraction intensities. As an example, we consider the normalized specular intensity given by

$$P_{00} = J_0^4[u_i(E)]. \tag{5}$$

In Table 1, the function $J_0^4(x)$ is tabulated for the range of values of x that we typically encounter in actual scattering processes. It is apparent that a small change in the argument may result in an appreciable change in the specular intensity. However, definite quantitative results cannot be given at this time because the manner in which the potential parameters D, α and β change from one electronic state to another is not known.

Let us now consider atom–surface scattering in the presence of laser radiation. We assume that the atoms are prepared in the ith electronic state and that the laser field consists of N photons. We also assume that the laser frequency is chosen to be

Table 1. Tabulation of $J_0^4(x)$ for the range of values of x typically encountered in an actual scattering process

x	$J_0^4(x)$	x	$J_0^4(x)$
0.6	0.6918	2.4	0.0000
0.8	0.5129	2.6	0.0001
1.0	0.3428	2.8	0.0012
1.2	0.2029	3.0	0.0046
1.4	0.1032	3.2	0.0105
1.6	0.0430	3.4	0.0176
1.8	0.0134	3.6	0.0236
2.0	0.0025	3.8	0.0263
2.2	0.0001	4.0	0.0249

resonant with the potential energy difference $V_{0_{i+1}} - V_{0_i}$ at a certain normal distance $Z = Z_r$. We take Z_r to be sufficiently large that the short-ranged potentials $V_{1_{i+1}}$ and V_{1_i} at $Z = Z_r$ are negligibly small; this means that we may safely consider the electronic transition and the diffractive transition to occur separately in time. For the electronic transition, we make the rotating wave approximation, considering only the two states $|i, N\rangle$ and $|i + 1, N - 1\rangle$, where $|i, N\rangle$ represents the state with the atom in the ith electronic state and with the radiation field having N photons. The potential energy curves for the two states are illustrated in Fig. 1. As the atoms move toward the surface, they come to the crossing point Z_r before diffraction occurs. Depending on the laser power and other details of the system, a certain fraction of the atoms makes a transition to $(i + 1)$th state. Those atoms then undergo diffractive scattering with the surface as if they were prepared in the $(i + 1)$th state with the incident energy $E + \Delta$. (See Fig. 1. This figure is drawn with the assumption that the crossing occurs with the laser frequency detuned to the blue by $\Delta/\hbar$. If the crossing occurs with the frequency detuned to the red by $\Delta/\hbar$, then $E + \Delta$ has to be replaced by $E - \Delta$.) The normalized diffraction intensities for those atoms are then given by

$$P_{nm} = J_m^2[u_{i+1}(E + \Delta)]J_n^2[u_{i+1}(E + \Delta)], \tag{6}$$

where $u_{i+1}(E + \Delta)$ is given by Eq. (3) with β_i, γ_i, D_i and E replaced by β_{i+1}, γ_{i+1}, D_{i+1} and $E + \Delta$, respectively. If the laser power is sufficiently high, then a large

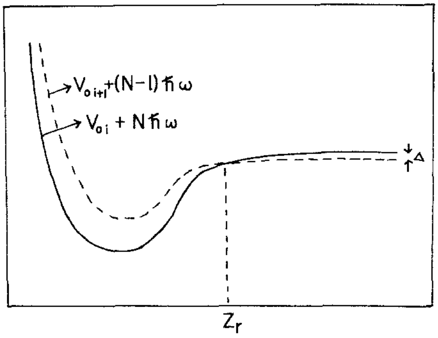

Fig. 1. Potential energy curves for the two states $|i, N\rangle$ and $|i + 1, N - 1\rangle$

fraction of the atoms may give diffraction patterns according to Eq. (6), and the effect of the laser radiation may be seen since the diffraction intensities predicted by Eq. (6) may be vastly different from those by Eq. (2). Judging from the results of the molecular scattering studies [2], we expect the laser power density to be $\sim MW/\mathrm{cm}^2$ or higher to produce an observable change in diffraction intensities.

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