Coalescent resonances in atom-surface collisions

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In atom-surface collisions, there are pairs of resonances which merge into a single resonance for special values of initial parameters of atomic beam. They display an unusual broadening effect when they are close to each other. In this paper we describe some of their properties based upon a model example.

I. INTRODUCTION

Resonances in atom-surface collisions have been thoroughly studied because they give very accurate information about the atom surface potential.[1–6] These resonances can be studied in different ways, depending on how their degrees of freedom are independently varied. Since there are three of these degrees of freedom, energy of the incoming atom, the azimuthal, and polar angles of its momentum with respect to the surface, there are a number of ways in which they can be independently varied. Usually two of them are fixed and the third is varied. For example, the azimuthal and polar angles are fixed and the collision energy is varied. In such a case the position of the resonance in the variable which is scanned is a function of the two degrees of freedom. In the preceding example it means that energy of the resonance is a function of the two angles. We will call these two degrees of freedom the "controlling parameters" because they determine the position of resonance in the third variable.

Let us look at a single resonance, for instance, in the energy variable. As the controlling parameters are changed, the resonance will "move" on the energy axis and in doing so it can come close to another resonance. Several things can happen then. Either they "repel" each other, cross without noticing each other, or they merge together. In the first case the effect is caused by the degenerate resonances, while in the second case the two resonances do not interact. The third case we will discuss in this paper and, as we will show, an unusual effect is observed. As the resonances approach each other, their width broadens until they merge together. After this point, the intensity of the merged resonance peak rapidly goes to zero. This property, as we will see, is something unique to these resonances, the "coalescent resonances" as we will call them.[7]

In this paper we will discuss two types of coalescent resonances: one in which the collision energy of atom is scanned and the other when the azimuthal angle is scanned (in this case the controlling parameters are the collision energy and the polar angle). We will also show their properties using a simple example.

However, before describing in detail these resonances, we will briefly review how they are located. We will assume the weak-coupling case so that perturbation theory can be applied.

The set of equations describing atom-surface scattering is[8]

$$\psi''_{\vec{G}} + K^2_{\vec{G}}\psi_{\vec{G}} = \sum_{G'} V_{\vec{G}-\vec{G}'}\psi_{\vec{G}'} , \tag{1.1}$$

where the channel energies $K^2_{\vec{G}}$ are

$$K^2_{\vec{G}} = k^2\cos^2\theta - G^2 - 2kG\sin\theta\cos(\phi-\phi_G) \tag{1.2}$$

and $\vec{G}$ is the inverse lattice vector. The angle ϕ_G is defined as

$$\tan\phi_G = \frac{G_y}{G_x} , \tag{1.3}$$

where G_x and G_y are the x and y components of $\vec{G}$ in the plane of surface. G in (1.2) is the module of $\vec{G}$. The angles θ and ϕ are the polar and azimuthal angles of the specular peak.

In the weak-coupling limit we solve the system (1.1) by neglecting the off-diagonal elements of the potential matrix, in which case (1.1) becomes

$$\psi''_{\vec{G}} + K^2_{\vec{G}}\psi_{\vec{G}} = V_{\vec{0}}\psi_{\vec{G}} . \tag{1.4}$$

In the uncoupled set of equations (1.4) we distinguish channels which are open ($K^2_{\vec{G}}>0$) and those which are closed ($K^2_G<0$). A resonance of the Feshbach-type is formed if one of the closed-channel energies coincide with a bound state of $V_{\vec{0}}$. Therefore an appropriate position of a resonance is obtained from the equation

$$-k_b^2 = k^2\cos^2\theta - G^2 - 2kG\sin\theta\cos(\phi-\phi_G) , \tag{1.5}$$

where $-k_b^2$ is the energy of a bound state of $V_{\vec{0}}$. Equation (1.5) implicitly gives the value of the position of the resonance in the variable which is scanned. For example, if collision energy is scanned, then (1.5) gives two values of k (k^2 is the collision energy in units of wave-number squared) for which resonance is observed. These values are

$$k_{1,2} = \frac{k_0^2 \pm [k_0^2-(k_b^2-G^2)\cos^2\theta]^{1/2}}{\cos^2\theta} , \tag{1.6}$$

where

$$k_0 = G \sin\theta \cos(\phi - \phi_G) \ . \tag{1.7}$$

The solutions of (1.6) are only meaningful if $k_{1,2}$ are positive, therefore not always will the two resonance peaks be observed for a given k_b^2 and G.

II. COALESCENT RESONANCES IN ENERGY

In the Introduction we described the source of the coalescent resonances and we also showed that there are several types of them, depending on which degree of freedom is scanned. In this section we will describe these resonances when k is scanned (or collision energy, which is k^2). As we mentioned, the position of the resonance in k is a function of the controlling parameters θ and ϕ. This relationship is explicitly given by (1.6). As the controlling parameters are changed, the position of the pair of resonances (1.6) will also change up to the point when $k_1 = k_2$. This will happen when

$$k_0 = \sqrt{k_b^2 - G^2} \cos\theta_c \tag{2.1}$$

or

$$\tan\theta_c \cos(\phi_c - \phi_G) = \frac{1}{G} \sqrt{k_b^2 - G^2} \tag{2.2}$$

if we use the definition (1.7). The index c of the two angles designates the specific values of these variables for which the two resonances (1.6) merge together. Equation (2.2) gives the value of one angle if another angle is arbitrarily specified. When θ_c and ϕ_c are obtained from (2.2), the collision energy of the merged resonances is $k^2 = k_0^2 / \cos^4\theta_c$.

However, in practice one would also want to specify the energy of the merged resonances and obtain θ_c and ϕ_c. This can be done if we combine (2.2) with (1.5) in which case we obtain

$$\begin{aligned} \cos\theta_c &= \frac{1}{k}(k_b^2 - G^2)^{1/2} \ , \\ \cos(\phi_c - \phi_G) &= \left[\frac{K_b^2}{G^2} - 1 \right]^{1/2} \cot(\theta_c) \ , \end{aligned} \tag{2.3}$$

where k^2 is the collision energy for which we want to observe the merged resonances.

The question now is how do these resonances affect the intensity of diffraction peaks? We will show this on a model example, which is general enough so that it incorporates all the essential elements for formation of these resonances. It should be pointed out that the essential properties of the coalescent resonances, and especially around the point where they merge together, are almost entirely determined by the structure of the channel energy and not the form of potential. After all, in the relationship (2.3) the potential only enters indirectly through the bound-state energies k_b^2. Therefore, the following model calculation will indeed be representative of the more general cases.

We will assume a model with the following five channels: $\vec{G} \equiv (m,n) = (0.0)$, (1,0), (0,1), $(-1,0)$, and $(0,-1)$, where (m,n) are the Miller indices. For diagonal elements $V_{\vec{0}}$ we will take

$$V_{\vec{0}} = \begin{cases} -19.28 \ \text{Å}^{-2} \ , & Z \le 2\text{Å} = Z_1 \\ 0 \ , & Z > Z_1 \end{cases} \tag{2.4}$$

while all the coupling matrix elements are

$$V_{\vec{G} - \vec{G}'} = \begin{cases} 1 \ \text{Å}^{-2} \ , & Z \le Z_1 \\ 0 \ , & Z > Z_1 \end{cases} \tag{2.5}$$

except the elements $\vec{G} - \vec{G}' = (2,0)$ and (0,2), which are zero. The potential V_0 has three bound states: $k_1^2 = -2.4077$, $k_2^2 = -11.4602$, and $k_3^2 = -17.2986$ Å^{-2}. For the lattice constant we have taken $a = 2.84$ Å.

The set of equations (1.1) can be solved in a closed[9] form for the potential (2.4) and (2.5). The appropriate S-matrix elements for the specular intensity is[6]

$$S_{\vec{0} \to \vec{0}} = \frac{f(-K_{\vec{0}})}{f(K)} \ , \tag{2.6}$$

where[9]

$$f(K) = e^{iz_1 K_{\vec{0}}} \det[Up \cot(zp) \tilde{U} - iK] \tag{2.7}$$

and

$$p = \tilde{U}(K^2 - V)U \ . \tag{2.8}$$

K^2 is the diagonal matrix of channel energies, while the matrix U diagonalizes $K^2 - V$, where V is the potential matrix. $f(-K_{\vec{0}})$ means that $K_{\vec{0}}$ in $f(K)$ is formally replaced by $-K_{\vec{0}}$.

Near the isolated resonance the S-matrix element (2.6) parametrizes as

$$S_{\vec{0} \to \vec{0}} \sim \frac{\beta}{k^2 - k_{\text{res}}^2} + b \ , \tag{2.9}$$

where k_{res}^2 is a pole of the S matrix, β is the residue, and b is the background term. The parametrization (2.9) will become useful in the analysis of the coalescent resonances. As it will be shown, the position of merged resonances is very sensitive to the change in θ and ϕ. However, when the coalescent resonances are relatively far from each other, this dependence is less sensitive. Therefore we can locate the appropriate poles of the coalescent resonances using (2.9), when they are separated, and follow the movement of these poles when θ and ϕ are varied. Using this procedure for locating the point of merger is much easier than following the resonance peaks. Furthermore, as it will be shown, the two poles of the coalescent resonances are located on the opposite sides of the real k^2 axis. Therefore we can easily identify resonances which are coalescent, namely, all the poles which correspond to the usual resonances are located in the lower half of the complex k^2 plane, while one of the two poles which correspond to the coalescent resonances, is in the upper half. Therefore, it is easy to identify this pole, while the identification of the other will immediately follow.

The simplest way to locate the approximate position where two coalescent resonances merge is by assuming a value of k for which this happens. Then we check whether for this value of k the equations (2.3) have solutions for certain $-k_b^2$ and G. If they do not have solutions, we must choose another k, but if they have, then the obtained values θ_c and ϕ_c give the approximate position of the merged resonances.

In our example we have taken $k=6.5$ Å^{-1}. For this value of k, the set of equations (2.3) has a solution for the bound state (0,1;2).[10] The values of θ_c and ϕ_c are

$$\theta_c=66.784^\circ\ ,\quad \phi_c=60.212^\circ\ . \tag{2.10}$$

Let us first analyze the poles of these resonances as a function of the controlling parameters. For simplicity we will assume that ϕ is fixed and equal to ϕ_c, while θ is varied. In Fig. 1 we show positions of the poles for typical values of θ. The poles were calculated by fitting (2.9) to the values of the S-matrix element (3.6) in the vicinity of a resonance.

Typical values of θ in Fig. 1 are indicated by the Roman numerals I—V, while the corresponding values of θ are $\theta=66.85^\circ$, 66.9°, 67°, 67.5°, and 68°, respectively. The cross in Fig. 1 indicates the approximate position of the merged resonances, given by (2.10). We notice that one pole is in the upper and the other in the lower half of the complex k^2 plane. For the values of θ away from θ_c the two poles are separated, which indicate that the corresponding resonance peaks are well separated. As the value of θ goes toward θ_c, the two poles acquire larger imaginary parts until the point where the change in θ causes only a change in the imaginary part of the pole, while the real part is practically unaffected.

In Fig. 2 we show how we observe these two resonances in the specular peak. We have calculated their positions for typical values of θ. As the parameter θ is changed, the two resonances come closer, while at the same time

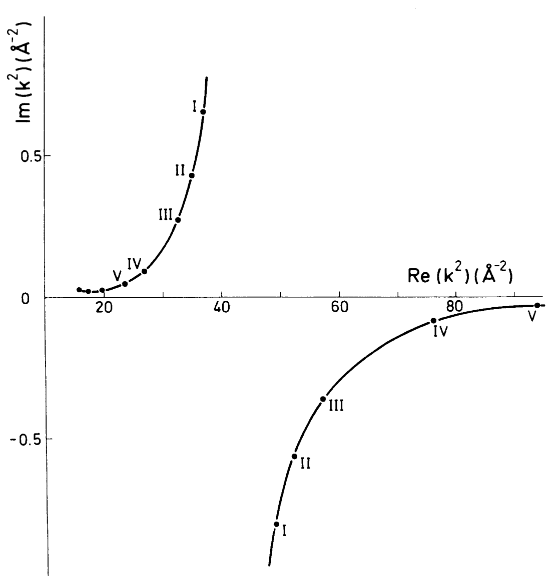

FIG. 1. Coalescent resonance poles for typical values of θ indicated by the Roman numerals. The values of θ are given in text.

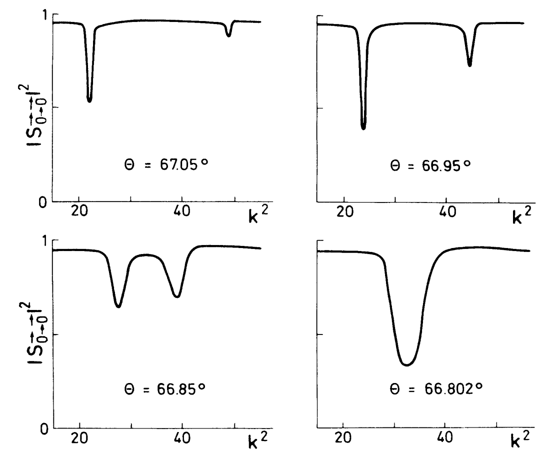

FIG. 2. How coalescent resonances appear in the specular peak. We notice the broadening effect described in text.

their width changes. At the point of merger, their width becomes large indeed and instead of two resonances we only observe one. However, the merger of two resonances is not real because their corresponding poles are well separated in the complex k^2 plane, but the real part of these poles is nearly the same. Nevertheless, we will call the merged resonance in Fig. 2 the "giant" resonance.

Simple behavior of the coalescent resonances in Fig. 2 is not always found. This is important to realize if one looks for them in other circumstances. We illustrate this with the example of the giant resonance for $k=7$ Å^{-1}, which has a source in the bound state (1,0;3). In such a case

$$\theta_c=59.793^\circ\ ,\quad \phi_c=22.060^\circ\ , \tag{2.11}$$

and the position of the poles which are found in the vicinity of the giant resonance are shown in Fig. 3.

The Roman numerals I—VII indicate the following set of angles θ: 60.1°, 60°, 59.9°, 59.8°, 59.7°, and 59.68°, and 59.35°, respectively. We notice a presence of a third pole, which has a source in the bound state (0,1;1). As the angle θ is varied, pole a in the upper half of the k^2 plane is

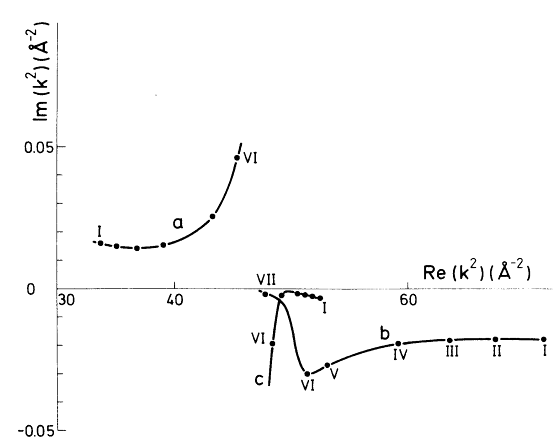

FIG. 3. Coalescent resonance poles when a third pole is between them.

practically unaffected by the presence of the third pole. Pole b initially corresponds to one of the coalescent poles, but as it approaches pole c an exchange effect occurs. The role of the coalescent pole is taken by pole c, while pole b replaces c. This effect is typical for the degenerate resonances.

In Fig. 4 we show how this effect is observed in the specular peak. Initially, the three resonances are well separated. As the resonance that is furthest right approaches the middle, it is "repelled" by it and stops its movement. The middle resonance takes the role of the coalescent resonance that is furthest right and forms the giant resonance with the left resonance. In search of a giant resonance, the exchange effect, which we have just described, may cause some difficulties.

III. COALESCENT RESONANCES IN ϕ

Instead of looking for the coalescent resonances in energy, we can study them in the variable θ. In such a case (1.5) also gives two solutions in ϕ, given by

$$\phi-\phi_G=\cos^{-1}\left[\frac{k^2\cos^2\theta-G^2+k_b^2}{2kG\sin\theta}\right] . \tag{3.1}$$

The two resonances will merge into a single one if

$$\frac{k^2\cos^2\theta-G^2+k}{2kG\sin\theta}=\pm 1 , \tag{3.2}$$

in which case ϕ is given by

$$\phi-\phi_G=0°, 180° . \tag{3.3}$$

The corresponding values of the controlling parameters when $\phi-\phi_G=0°$ are

$$k_c=\frac{G\sin\theta_c\pm(G^2-k_b^2\cos^2\theta_c)^{1/2}}{\cos^2\theta_c} , \tag{3.4}$$

where we have assumed that θ_c is arbitrary. We notice that there are two values of k for which we observe the giant resonance, provided both values of k are positive.

Similarly, when $\phi-\phi_G=180°$ we obtain

$$k_c=\frac{-G\sin\theta_c+(G^2-k_b^2\cos^2\theta_c)^{1/2}}{\cos^2\theta_c} \tag{3.5}$$

and for only one value of k the giant resonance is observed (the other value of k_c is always negative).

An interesting effect is observed when $\phi-\phi_G=0°$. We noticed from (3.4) that there are two values of k_c for which the giant resonances are observed. These two giant resonances can also merge into a single one if

$$G=k_b\cos\theta_c . \tag{3.6}$$

We can call such a resonance the "supergiant" resonance and its position is given by

$$\cos\theta_s=\frac{G}{k_b} , \quad k_s=\frac{k_b}{G}\sqrt{k_b^2-G^2} , \tag{3.7}$$

where $\phi-\phi_B=0°$.

We have calculated two examples of the coalescent resonances in the ϕ variable. In the first we show formation of a giant resonance and in the second the supergiant resonance. For the position of the giant resonance we have taken $\phi=90°$, since this is arbitrary, and $\theta_c=60°$ which is also arbitrary. It follows that for the bound state (0,1;2) k_c is given by (3.5) and we have taken the larger of the two values, $k_c=13.363$ Å^{-1}. For the value of θ_c we have calculated the poles of the S matrix in the variable ϕ for different values of k^2. They were calculated by fitting the S-matrix element (2.6) near a resonance by the function

$$S_{\vec{0}\to\vec{0}}=\frac{\beta}{\phi-\phi_p}+b , \tag{3.8}$$

where ϕ_p is complex. The results of calculation are shown in Fig. 5.

Typical pole positions are shown by the Roman numerals I–VIII for the following values of k^2: 175, 176, 177, 178, 178.5, 178.8, 179, and 179.02 Å^{-2}, respectively. In Fig. 6 we show how these resonances appear in the specular peak.

Their behavior is very similar to the coalescent resonances in k^2. However, it does not mean that the giant resonance in Fig. 6 will also be observed if ϕ is fixed to

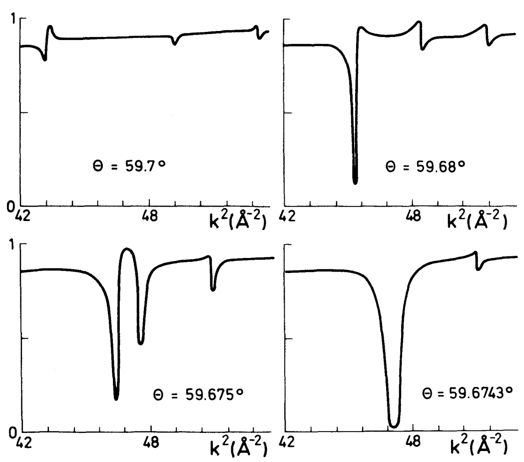

FIG. 4. Behavior of the coalescent resonances when an exchange effect occurs with a third resonance.

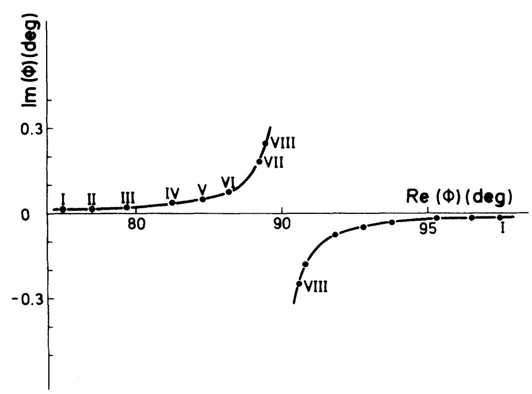

FIG. 5. Coalescent resonance poles in the ϕ variable. Roman numerals indicate different values of k^2 given in text.

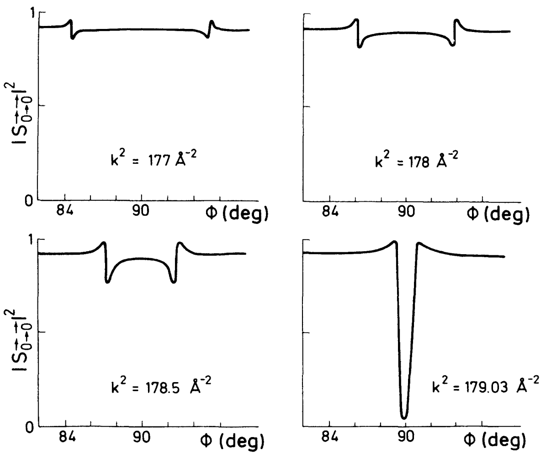

FIG. 6. How coalescent resonances in ϕ appear in the specular peak.

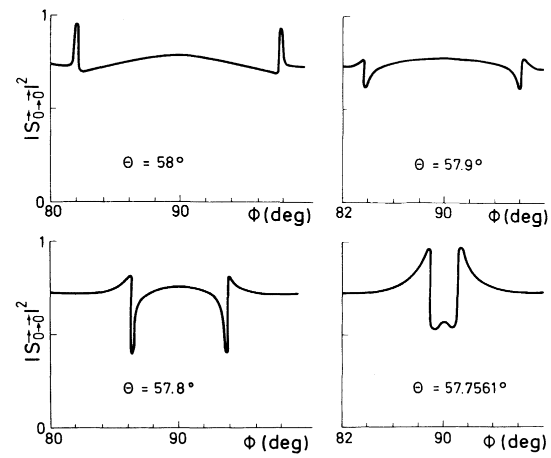

FIG. 7. Formation of a supergiant resonance.

90°, θ has the value of 60°, and k^2 is scanned around $k^2=179.03$ Å^{-2}. The property of a giant resonance is that it can only be observed if the appropriate variable is scanned. It cannot be observed when we scan all variables. This property follows from the fact that although the conditions (3.3) and (3.4) are satisfied for the giant resonance in ϕ, they do not satisfy the conditions (2.3) for the giant resonance in k^2. The exception is the supergiant resonance. Its formation is shown in Fig. 7 for the bound state (0,1;3), $\theta_s=57.864°$, and $k_s^2=43.838$ Å^{-2}. One can easily show that this resonance is observed in both the ϕ and k^2 variables.

IV. CONCLUSION

In this paper we have shown properties of a pair of resonances which we called the coalescent resonances. They are observed when one of the parameters, which determine the initial condition of the incoming atom, is scanned. Under the special circumstances discussed in this paper these two resonances form a single resonance of very large width and we called them the giant and supergiant resonances. The giant and supergiant resonances are only superficially single resonances because, as we have seen, they are represented by two poles of the S matrix of almost equal real parts. We did not study these resonances when θ is scanned, but it is believed that no further insight would be gained when this is done.

ACKNOWLEDGMENT

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[10]We use the standard notation for the bound states in the atom-surface system. The numbers $(m,n;b)$ designate two Miller indices and the vibrational quantum number.