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# Vacancy-mediated diffusion of Co atoms embedded in Cu(001)

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ABSTRACT

The diffusion of Co atoms in the Cu(001) surface has been studied using Scanning Tunneling Microscopy (STM). Like other impurities in the Cu(001) surface, the diffusion of Co is mediated by single surface vacancies. STM images reveal that diffusion of the embedded atoms takes place through multi-atom jumps separated by long time intervals, which is characteristic for this type of diffusion. The jump length and frequency are measured to establish the nature of the interaction between surface vacancies and the embedded Co atoms and to extract the relevant formation and diffusion energies.



## 1. Introduction

Thin magnetic films grown on non-magnetic substrates nowadays find application in various magneto-electronic devices. Magnetic thin films exhibit important interface phenomena such as interlayer exchange coupling, giant magnetoresistance [1] and surface anisotropy [2,3]. The magnetic properties of the films are directly related to the interface quality. This makes the degree of alloying that occurs between a magnetic adlayer and substrate in the initial stages of growth of a film of fundamental interest. Studies of metal growth have been performed on several fcc(100) metal surfaces for which the alloy formation in the bulk is known, such as Rh/Ag(100) [4], Au/Cu(100) [5–7] and Pd/Cu(100) [8]. In some cases, such as for Au/Cu(100) [5–7], stable ordered surface structures are imaged with STM that are related to a known bulk phase. For metal-substrate combinations that are bulk immiscible, intermixing is often confined to the first atomic layer of the substrate [9]. These heteroepitaxial systems form surface alloys by exchanging deposited atoms with substrate atoms to lower the total free energy of the system.

One such system, which has been studied both experimentally and theoretically, is Co on Cu(001). In the low coverage regime (<0.1 ML) and for temperatures slightly above room temperature, deposited Co atoms exchange positions with Cu terrace atoms. Three diffusion mechanisms are active: those for Cu and Co adatom diffusion and the one for embedded Co diffusion. Already for simple homoepitaxial systems, diffusion of individual atoms can be quite complicated [10,11]. Here, we expose and quantitatively analyze the diffusion mechanism that allows Co atoms to diffuse whilst remaining embedded in the first layer of the Cu(001) substrate.

## 2. Experimental

A low coverage of Co (<0.08 ML) was deposited at less than 1 ML/min in a temperature range of 330 K to 380 K. Prior to deposition the surface has been cleaned by repeated cycles of $Ar^+$ bombardment and annealing. The purity of the surface has been verified with STM and revealed atomically clean terraces with an average width of 1000 Å. A Co–Cu exchange mechanism is active at this temperature and Co atoms immediately exchange with Cu surface atoms after deposition to form a surface alloy. The Co atoms are imaged as protrusions with a typical corrugation of the order of 0.1 Å in STM images, that were acquired with bias voltages in the range of −2.5 V to +2.5 V. The embedded Co atoms have a tendency to form clusters in Cu(001) after deposition, as illustrated in Fig. 1(a). In the experiments that are detailed in this manuscript we have focused exclusively on isolated Co atoms. After Co deposition the sample temperature is lowered to the temperature of the experiment. To quantitatively analyze the motion of individual embedded atoms, the position of the Co was measured from STM image sequences, recorded at 294 K and 317 K. A stable sample temperature and an equilibration in temperature between STM and sample were a prerequisite to obtain a minimum in thermal drift. Further drift effects were eliminated by measuring the Co diffusion relative to fixed reference points in consecutive images. These reference points, generally embedded Co atom clusters with more than three atoms, allowed us to further correct for distortions in the x and y directions between

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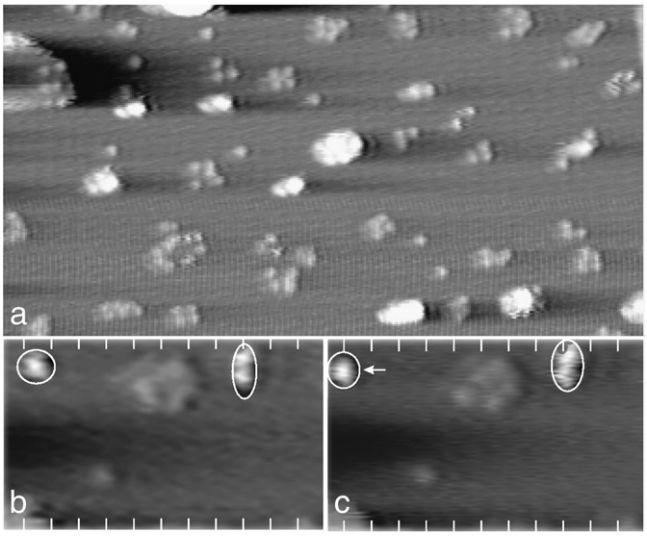


**Fig. 1.** (a) A $162\times84$ Å$^2$ STM image of the Cu(001) surface directly after deposition of the Co. Embedded Co clusters and individually embedded Co atoms can be seen as protrusions in the image. Between (b) and (c) the embedded atom indicated by the arrow makes a diagonal jump with a total length of $\sqrt{2}$ lattice spacing. The time elapsed between images is 38 s and the sample temperature was 317 K. The image size is $64\times38$ Å$^2$, $I_t=0.7$ nA, $V_t=2.31$ V. The distance between the two white lines is 5.4 Å and the fuzzy appearance of the features is the result of mechanical noise coupling to the instrument.

images. Only those embedded Co atoms that were sufficiently far away, i.e. at least two times the length of the largest long jump observed, from embedded clusters or large Co decorated Cu islands, were considered for our analysis. If an influence of these clusters exists, it can therefore be safely neglected. Due to these self-imposed constraints, the total data set is however also limited, nevertheless sufficiently large to be able to extract an accurate value for the diffusion barrier. The acquisition time of all images ranged between 30 and 240 s, which was sufficient to observe the diffusion of the embedded Co atoms, as is illustrated in Fig. 1.

## 3. Possible diffusion mechanisms

After deposition at room temperature, Co atoms are immediately incorporated into the Cu(001) surface through an exchange mechanism. For low coverages the embedded Co atoms diffuse and may form small clusters in the substrate. Several mechanisms through which the embedded atoms can diffuse are known from literature. They may diffuse via the mediation of a Cu adatom; here the Co atom is visited by a Cu adatom, as shown in Fig. 2. A Cu adatom first exchanges places (panels (a) and (b)), then the newly created Co adatom diffuses on the Cu surface (panel (c)), before it reincorporates into the surface via a new exchange process (panel (d)). This adatom mediated diffusion was found to be active for a Cu/Co monolayer on Ru(0001), by Schmid et al. [12].

A second possibility for an embedded Co atom to diffuse is through vacancy-assisted motion. In this mechanism, a single surface vacancy which diffuses rapidly through the first layer of the substrate, is the mediating particle. A vacancy approaches the embedded Co atom from any of the four possible <110> directions, as illustrated in Fig. 3. After the vacancy arrives (panel (a)), it changes places with the Co atom (panels (b) and (c)), and ultimately leaves the Co atom displaced (panel (d)). A frequent visit by one and the same vacancy allows the embedded Co atom to be displaced over a distance of several lattice spacing. The diffusion of Mn [13], In [14], Pd [15] and Pb [16,17] embedded in the Cu(001) surface has been ascribed to the vacancy exchange mechanism.

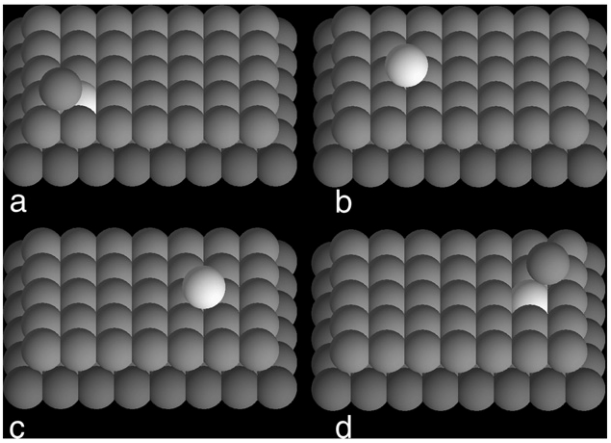


**Fig. 2.** Adatom mediated diffusion: Embedded Co is visited by a Cu adatom (a) and exchanges places (b). Once on top, the Co adatom diffuses over the surface (c) and then exchanges back with a substrate atom (d). For Co atoms that move to the original, NN (nearest-neighbor) or NNN (next-nearest-neighbor) positions, no diffusion over the surface is necessary, only exchanges.

A third and final diffusion mechanism that may be considered is diffusion that is aided by an adsorbate, where the adsorbate forms a complex with the embedded atom when it arrives from the gas phase. So far, no study is known which investigates the diffusion of embedded atoms mediated by an adsorbate on a (001) surface. For (110) surfaces, both adatom [18] and vacancy [19] mediated diffusion via adsorbates is known to occur. This particular mechanism is however not able to explain the collective occurrence of long jumps that is observed in this system, where many atoms make long jumps simultaneously. With this background information, we now interpret the acquired data.

### 3.1. Jump length distribution

Our data set consists of the distribution of *long jumps* and the frequency with which those long jumps occur. Consecutive jumps are separated by long time intervals, determined by the time for a new vacancy to arrive at the embedded atom. In the mechanisms discussed in the previous paragraph, the displacement of the adatom or vacancy is assumed to take place via a burst of single hops between adjacent sites. For all three mechanisms, the STM is not able to observe the individual moves that constitute one long jump. However, it can determine the probability of finding the particle at a specific location after the long

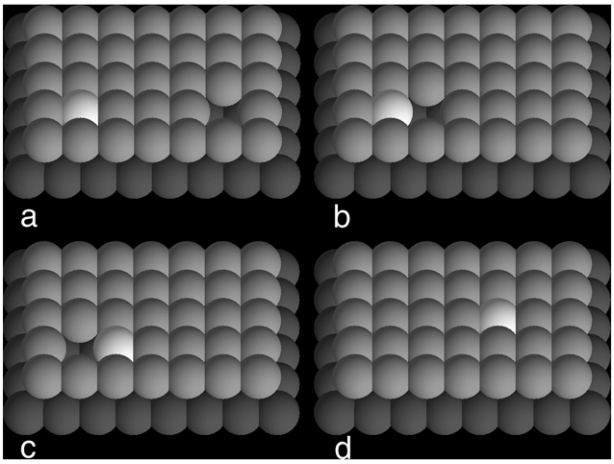


**Fig. 3.** Vacancy mediated diffusion: Embedded Co is approached by a vacancy from one of four possible directions (a,b), and changes place with the vacancy to move one lattice spacing (b,c). A frequent visit allows the Co to move over several lattice spacings.

jump has taken place by closely examining the distribution of long jump lengths. For all three mechanisms the random walk of the diffusing particle is suddenly terminated by either the de-excitation of the adatom, the exchange of the diffusing adatom or the annihilation of the vacancy. This sudden termination of the random walk leads to a peculiar shape of the distribution of jump lengths. For this type of random walk, the distribution of jump lengths has been shown to have the shape of a modified Bessel function [20–22]. For vacancy mediated diffusion in two dimensions this is the modified Bessel function of zeroth order as was established in Ref. [14]. The probability for a jump of length r in a time interval $t$ is given by

$$P_t(\mathbf{r}) = \frac{2(\pi-1)}{\log t} K_0\left(\frac{r}{[\log t/(4\pi(\pi-1))]^{\frac{1}{2}}}\right) \quad (1)$$

Under the assumption that the individual exchanges of the vacancy and the embedded atom are uncorrelated, i.e. there is no significant interaction present, this Bessel function can be derived from the effective diffusion equation that is given by [14]

$$\frac{\partial \varrho(r,\tau)}{\partial \tau} = D_{eff}\nabla^2\varrho - c\varrho \quad (2)$$

where the quantity $c$ in the second term on the right hand side accounts for the probability that the vacancy is annihilated and the random walk is terminated. The quantity $\tau$ is the time interval between consecutive encounters of the diffusing vacancy and the embedded atom and $\varrho(r,\tau)$ denotes the probability to find the embedded Co atom at position $r$ after a time $\tau$. This implies that the diffusion coefficient in Eq. (2) is an effective diffusion coefficient that cannot be easily compared to the tracer diffusion coefficient that describes thermally activated single particle diffusion. The width of the jump length distribution is to a large extent determined by the interaction between the diffusing vacancy and the embedded atom. If the vacancy and the embedded atom repel each other, place exchange will be difficult and few exchanges will take place in a long jump. As a result the distribution of jump lengths will be narrow. On the contrary, if a strong attraction exists between the vacancy and the embedded atom, the two will behave like a quasi-bound pair, resulting in a great number of exchanges, which in turn will lead to a rather wide jump length distribution. The interaction between the vacancy and the embedded atom also affects the rate at which long jumps take place. Assuming Arrhenius-like behavior and no interaction between the vacancy and the embedded atom, the barrier that the impurity atom has to overcome to make a long jump is the sum of the formation energy of a vacancy ($E_{vf}$) and the vacancy diffusion barrier ($E_{vd}$). It is thus important to realize that the diffusion barrier for a long jump is an effective barrier given by

$$E_{eff} = E_{vf} + E_{vd}. \quad (3)$$

This simple picture where the diffusion of an embedded atom through long jumps is governed by the sum of the vacancy formation energy and the vacancy diffusion barrier changes when an interaction is present as will be discussed below. If the STM measurements are performed with a finite temporal resolution and if an interaction exists between the tracer atom and the vacancy, the effective barrier changes, as is described in [20]. In summary, the hopping rate and effective diffusion barriers are changed as found in Table 1.

## 4. Results and discussion

To establish the single particle diffusion coefficient and determine the effective diffusion barrier $E_{eff}$ we measured the mean square displacement of an embedded Co atom as a function of time, see Fig. 4. For two independent measurements performed at 317 K and 294 K, we find an effective diffusion barrier of $0.90 \pm 0.02$ eV and $0.91 \pm 0.02$ eV, respectively. To obtain these energy values we have assumed a prefactor of $10^{13}$ Hz [22]. If we assume that the diffusion is vacancy mediated (as will be shown in what follows), and if we assume that there is no interaction between the vacancy and the Co atom, then using $E_{eff}$ and a vacancy formation energy of 0.474 eV [15], we find a vacancy diffusion energy of 0.44 eV.

**Table 1**
Change of hopping rate and effective diffusion barrier as function of interaction energy between impurity atom and vacancy in a lattice of L×L, from [20], where $0 \le C \le 1$ is a constant determined by the geometry of the lattice, $E_{xd}$ represents the exchange barrier between the impurity atom and the vacancy and $\nu_{Lj}$ is the rate of the long jumps.

| Interaction | Hopping rate | Effective barrier |
|---|---|---|
| Identical to Cu atom | $\nu_{Lj} = \frac{\nu_0}{1+\frac{1}{3(1-C)}}$ | $E_{vf}+E_{vd}$ |
| Extremely attractive | $\nu_{Lj} = 3\nu_o(1-C)$ | $E_{vf}+E_{vd}$ |
| Extremely repulsive | $\nu_{Lj} = \nu_0$ | $E_{vf}+E_{xd}$ |

To properly establish the type of diffusion mechanism, we need to evaluate how the embedded Co atoms move and the diffusion statistics have to be analyzed in more detail. Following Ref. [23], a displacement distribution was measured. This distribution is the probability that an atom has moved a certain distance in one long

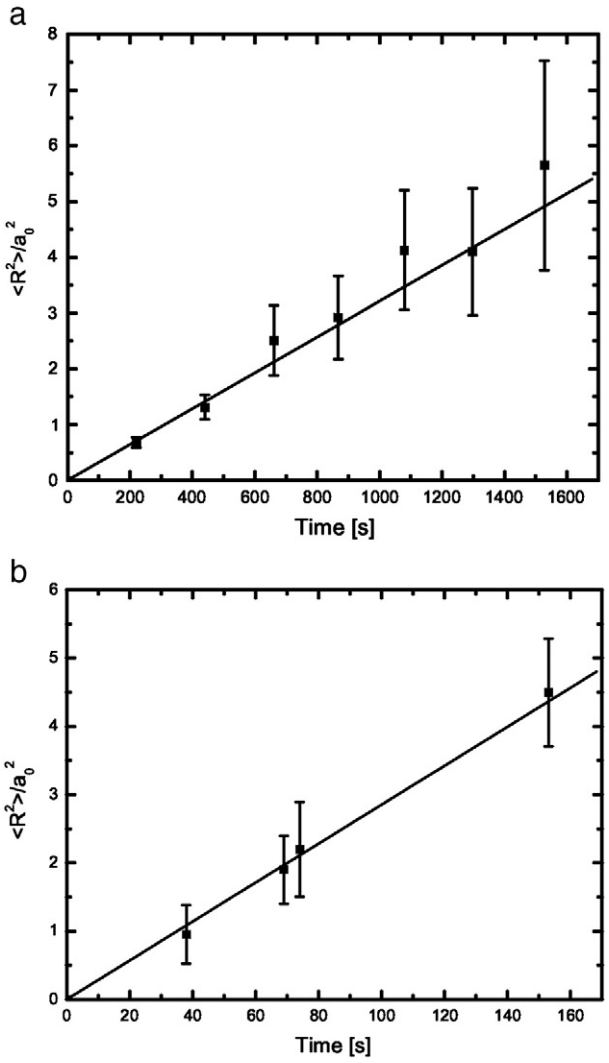


**Fig. 4.** Mean square displacement of embedded Co atoms versus time, measured at 294 K and 317 K.

jump and can be considered as a blue print for the type of diffusion process that takes place. By considering the fraction of jumps of length $\sqrt{2}a$, following Grant et al. [15], one can discriminate between adatom mediated diffusion and vacancy mediated diffusion. The essence of the argument is that for the adatom exchange mechanism to be viable, the probability for diffusion and exchange have to be approximately identical, which results in a rather simple, statistically determined jump length distribution with a probability for $\sqrt{a}$ jumps that is exactly half the probability of jumps of unit length. In analyzing the jump length distribution, see Fig. 5, we measure a ratio of $\sqrt{2}a$ hops to single hops of approximately 0.2, which is significantly lower than the number of 0.5, which is expected for the adatom exchange mechanism. Thus the diffusion of embedded Co atoms in a Cu(001) surface is mediated by vacancies. Furthermore, when comparing the statistics of Ref. [15] to those in our Co experiments, we find that the two are almost a carbon copy, see Table 2. The similarity goes further, since both Pd [15] and Co exchange immediately with a Cu surface atom after deposition, while In [20,24,25] and Pb [16] prefer to diffuse into the terrace via a step. Another similarity is that both embedded Pd and Co atoms are imaged as protrusions on the surface. Finally, the activation energies that were measured for both systems are very similar. For Pd/Cu(001) an activation barrier of 0.88 eV was measured, while we obtained an activation barrier of $0.91 \pm 0.02$ eV. In view of these similarities we expect the interaction between a Co atom and a vacancy to be moderately repulsive, as was established for the Pd atoms [15].

The fact that we are dealing with a repulsive interaction can also be established from the width of the jump length distribution. It determines the type of interaction that exists between the Co atom and the vacancy [23]. If we compare the width of our jump length distribution, which is on the order of 1 NN spacing, with the width of the jump length distribution that was obtained for e.g. In, we find that the distribution that we measured for Co is significantly narrower, indicating a significantly more repulsive interaction than existed for In. The repulsive interaction that apparently exists for Co decreases the number of moves that a Co atom will make in one long jump, since the vacancy will find it more difficult to approach the Co atom. The probability for long jumps of a large length will be reduced as can also be seen from the jump length distribution that was found for Pd, where after a distance of 3 NN spacings the jump length distribution is practically zero [15]. In fact, intuitively we expect a repulsive interaction since the Co atom exchanges places with a Cu atom

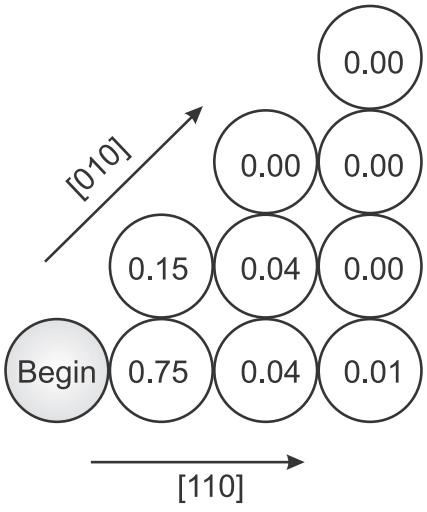


**Fig. 5.** The distribution of jump vectors measured from 68 jumps that were observed in STM movies at 300 K. Plotted is the probability for jumps of a Co atom from its starting position to each of the lattice sites shown. The jump length of zero cannot be measured with STM and has therefore been ignored in the distribution.

**Table 2**
Comparison of the probabilities found for different hop lengths for the Pd/Cu(001) and Co/Cu(001) systems.

| Hop Length (lattice constants) | Pd/Cu(001) | Co/Cu(001) |
|---|---|---|
| 1 | 0.75 | $0.75 \pm 0.10$ |
| $\sqrt{2}$ | 0.17 | $0.15 \pm 0.05$ |
| 2 | 0.043 | $0.04 \pm 0.02$ |
| $\sqrt{5}$ | 0.026 | $0.04 \pm 0.02$ |
| 3 | 0.001 | $0.01 \pm 0.01$ |

immediately after deposition. This indicates that it is energetically more favorable for the Co atom to be surrounded by four Cu atoms. A vacancy that occupies one of the nearest neighbor positions would therefore reduce the coordination and increase the total free energy of the configuration.

Having established that the nature of the interaction between the vacancy and the embedded Co atom is repulsive, we re-evaluate the value of the activation energy that we obtained previously. It was shown in Ref. [20] that in the presence of a repulsive interaction between a vacancy and the embedded atom, the interpretation of the effective diffusion barrier changes and is given by (see Table 1)

$$E_{eff} = E_{vf} + E_{xd} \tag{4}$$

The measured activation energy is thus the sum of the vacancy formation energy and the Co vacancy exchange barrier. Assuming a value of 0.474 eV for the vacancy formation energy [15], we obtain a value of 0.44 eV for the Co vacancy exchange barrier. From this we conclude that the effective repulsive interaction must be somewhat smaller than what was found for Pd, as the exchange barrier tends to be closer to the diffusion barrier for a vacancy in clean Cu. To quantitatively verify the value of the interaction energy that we obtained, we now compare our experimental results with numerical calculations.

## 5. Calculations

To quantitatively determine the interaction energy between the vacancy and the embedded Co atom we perform a simple numerical calculation. By varying the interaction energies between the Co atom and the vacancy in NN positions, a jump length distribution is calculated, which is then fitted to our measured distribution via a normalized $\chi^2$ test , which assesses the goodness of the fit. Through this fitting procedure and with the experimentally measured terrace width, an accurate value for the interaction energy can be determined. To calculate a jump length distribution we employed the same discrete model that has been used to describe the In/Cu(001) vacancy mediated diffusion [20]. The model consists of a two-dimensional square lattice of finite size $(L \times L)$, which is centered around the origin and represents the top most Cu(001) layer. In this lattice all positions are occupied by Cu atoms, except for the impurity atom (i.e. for our case Co) and the vacancy. They are located at the origin (0,0) and at the first NN position in the positive x direction (1,0), respectively. This state corresponds to the configuration of the Co and the vacancy in the terrace after they have made an initial exchange. This initial exchange is a prerequisite for the observation of a long jump in the STM images and thus represents a proper starting configuration for the numerical calculations. The vacancy moves through the surface by performing a random walk. This random walk includes exchanges with the impurity Co atom. The random walk is terminated at the moment that the vacancy hits a boundary. In reality, this annihilation corresponds to the recombination of the vacancy with the step-edge. The random walk that the vacancy performs is biased since the exchange rate for each direction depends on the local environment, i.e. the rates with which the vacancy moves in each of the four possible directions are determined by the relative position that the

vacancy occupies with respect to the impurity atom, schematically depicted in Fig. 6. The further the vacancy is away from the impurity atom, the less the exchange rates are influenced by the impurity Co atom. In fact, in the case of a repulsive interaction, the diffusivity of the vacancy in the vicinity is adequately described by solely including a modified barrier for diffusion process 1 of Fig. 6. The long jump that the impurity atom makes is the result of all the exchanges that were made during the random walk of one individual vacancy. To find the displacement distribution of the impurity atom, we average many different vacancy random walks. We note that this distribution cannot be determined analytically due to the exchange rate distribution and the finite lattice size. The general method applied in Ref. [20] leaves only two fitting parameters: the lattice size and the value of the modified barrier versus $k_BT$. The lattice size is known quite well from our STM measurements and is approximately equal to 400 lattice spacings or 1000 Å.

This leaves us with the modified barrier as the only fitting parameter. In the numerical calculations we varied the modified barrier around the standard diffusion barrier of a vacancy in Cu(001) (0.426 eV [15]), plus an additional 25 meV repulsive interaction. The jump length distributions that we obtained for each of the modified barriers were compared with the measured jump length distribution. The results of this comparison are given in Fig. 7. It shows the normalized $\chi^2$ values that were obtained for each of the calculated distributions. The amount of data in the measured jump length distribution did limit to some extent the $\chi^2$ test, however, guiding values were obtained. From Fig. 7 we observe that the minimum $\chi^2$ value shifts to lower values of $E_{eff}/k_BT$ with decreasing terrace size. This is not unexpected since a narrow terrace will automatically shorten the random walk of the vacancy and lead to fewer exchanges of the vacancy and the impurity atom. Or, in other words, to obtain an equally good fit on a larger terrace, the repulsive interaction between the vacancy and the impurity atom needs to be increased to reduce the number of exchanges between them. For the experimentally observed terrace width of ≈1000 Å, we find a best fit repulsive interaction energy between the embedded Co atom and the vacancy of $36 \pm 13$ meV at room temperature. The jump length distribution that we obtained for this interaction energy is shown in Fig. 8. The best-fit value agrees well with the 40 meV interaction that was found for Pd/Cu(001) [15].

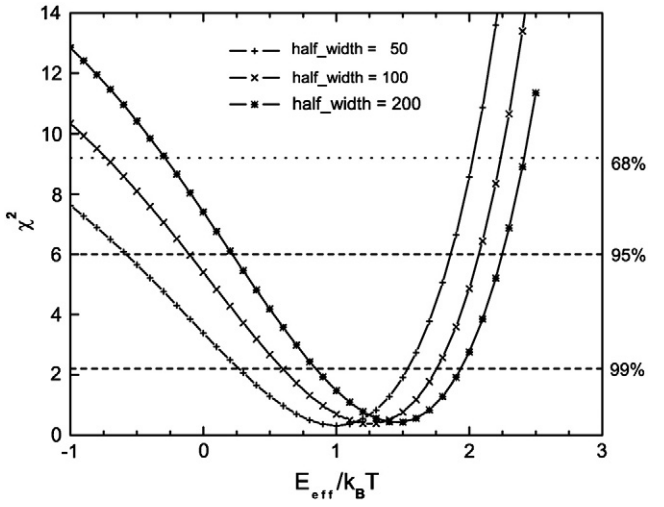


**Fig. 7.** Normalized $\chi^2$ values obtained for fits of the measured jump length distribution for different values of the modified diffusion barrier and lattice size. Dashed lines indicate the $\sigma$, $2\sigma$ and $3\sigma$ values for a Gaussian variable.

## 6. Conclusion

We have investigated the diffusion mechanism of embedded Co in a Cu(001) surface. Through our STM measurements, we find that embedded Co moves through the surface via a vacancy-mediated diffusion mechanism. The vacancy-mediated diffusion mechanism for impurities embedded in fcc(001) metal surfaces appears to be a general phenomenon. For the case of Co this was established by measuring the distribution of jump lengths of the Co atoms and comparing its shape to what is expected for both adatom- and vacancy-mediated diffusion. By evaluating the diffusion of embedded Co atoms, we measured the activation energy for the diffusion of Co in Cu(001). For this activation energy we find a value of $0.91 \pm 0.02$ eV. This number is similar to the number that was obtained for Pd/Cu(001) and indicates a moderate repulsive interaction between the vacancy and the embedded Co. We verified the value of the activation energy and the interaction that exists by comparing the experimental results with numerical calculations. We were able to fit the measured jump length distribution and obtain a value for the Co-vacancy exchange barrier. The value of 0.44 eV that we find is in excellent agreement with the value that was measured, and also with the value that was previously obtained for the case of Pd [15]. The similarity between these two systems and the repulsive interaction that is found for both Co and Pd, is assumed to be a result of the smaller atomic radius of both elements [26] compared to e.g. In or Pb. This will in turn affect their ability to relief the tensile stress that is present in the Cu (001) surface and cause them to interact differently with vacancy defects than other embedded atoms.

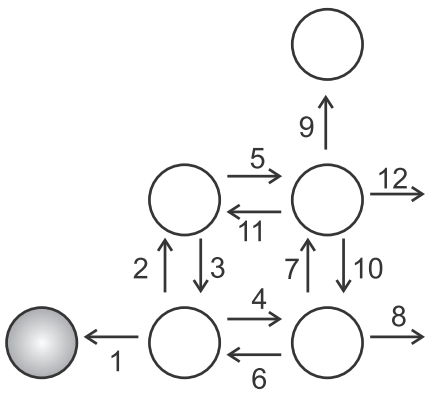


**Fig. 6.** Diffusion barriers for a vacancy near a Co atom.

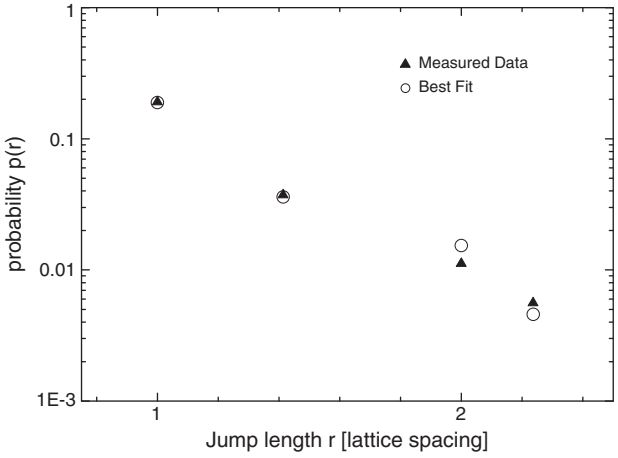


**Fig. 8.** Best fit jump length distribution for the measured data. Probabilities have been normalized to add up to one for the entire lattice, i.e. not exclusively for the non-equivalent lattice sites.

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