Role of the induced dipole moment in the collisional interference in the pure rotational spectrum of HD-He and HD-Ar

B. McQuarrie and G. C. Tabisz

Department of Physics, University of Manitoba, Winnipeg, Manitoba, Canada R3T 2N2

B. Gao

Department of Physics and Astronomy, University of Toledo, Toledo, Ohio 43606

J. Cooper

Joint Institute for Laboratory Astrophysics, University of Colorado and National Institute of Standards and Technology, Boulder, Colorado 80309-0440

The parameters describing the interference between allowed and collision-induced transitions in the far-infrared spectrum of HD-He and HD-Ar are computed following the formalism of Gao et al. [Phys. Rev. A **44**, 7379 (1991)] and the calculational procedure of Gao et al. [Phys. Rev. A **46**, 5781 (1992)]. The contribution from each component of the induced-dipole moment is identified. The interference between the induced-dipole component having the same symmetry as the allowed moment and the allowed moment itself has the largest single contribution to the intensity interference effect. The mixing of rotational levels by an anisotropic intermolecular interaction introduces other components into the interference mechanism. The overall effect is usually to decrease the magnitude of the interference and to permit line broadening and asymmetry to occur. This interaction is most effective in causing the anisotropic overlap-induced pair dipole moment $B_2(21)$ to play a role in the interference mechanism. The effect of the isotropic overlap-induced dipole moment is usually negligible.

I. INTRODUCTION

The small permanent dipole moment of the hydrogen isotopomer HD makes it an exceptionally well-suited molecule for the study of interference effects between allowed and collision-induced transitions. A considerable amount of experimental data have been amassed for the pure rotational transitions of HD in inert gas mixtures [1–3]. The spectral features of interest are sharp lines appearing at the frequencies of the purely allowed transitions and sitting on a broad collision-induced background. The intensities of these lines do not behave as do those for purely allowed transitions, but show an anomalous density and temperature dependence. This behavior can be associated with the occurrence of interference between allowed and induced transitions [4–6].

A general theory of the phenomenon has been developed which contains provision for allowed, purely induced, and interference (cross-term) contributions to the absorption coefficient within the binary collision approximation. Effects due to m-mixing, J-mixing, and succeeding, but not necessarily successive, collisions are taken into account. Calculations based on this formalism have been performed for the HD-He and HD-Ar systems and compared with experiment [8]. The results demonstrate the crucial importance of inelastic rotational collisions to the interference and line broadening mechanisms.

The present paper revisits those calculations. The identification and modeling of the various components of the induced dipole moment have formed a critical theme in the development of understanding of induced spectral transitions [9]. The purpose of the present study is to point out how aspects of the interference process are sensitive to details of the induction interaction.

II. THEORY

The theory is fully explained in Refs. [7] and [8]. Only a brief review of the essential expressions is given here. The absorption coefficient $\alpha(\omega)$ for a transition from an initial state J_g to an excited state J_e is

$$\alpha(\omega) = n_R \left[\frac{4\pi\omega}{3\hbar c} \right] (1 - e^{-\beta\hbar\omega}) P(J_g) \mathrm{Re}[i(\Delta + i\gamma)^{-1}(\mu_R + n_p\mu_1)(\mu_R + n_p\mu_2)] , \tag{1}$$

where $\beta = 1/k_B T$; ω is the frequency of the incident light, ω_{eg} is the molecular transition frequency, $\Delta = \omega - \omega_{eg}$ is the detuning, and $P(J)$ is the normalized Boltzmann distribution. The complex line broadening cross section γ is given by

$$\gamma = n_p \left[1 - \sum_{\substack{m_e,q,m_g \\ m_e',m_g'}} (-1)^{m_e'+m_e} \begin{bmatrix} J_e & 1 & J_g \\ -m_e' & q & m_g' \end{bmatrix} \begin{bmatrix} J_e & 1 & J_g \\ -m_e & q & m_g \end{bmatrix} \langle J_e m_e' | \hat{S} | J_e m_e \rangle \langle J_g m_g' | \hat{S} | J_g m_g \rangle^* \right]_a , \tag{2}$$

where $\hat{S}$ is the S matrix for the collision between the radiator R (HD molecule here) and the perturber (He or Ar atom).

$$\mu_R = \langle J_e \| \hat{\mu}_R \| J_g \rangle \tag{3}$$

is the reduced matrix element for the allowed radiator dipole. The n_p and n_R are perturber and radiator number densities. The dipole moments μ_1 and μ_2 are given in terms of the collision-induced dipole operator $\hat{\mu}_e$ by

$$\mu_1 = \sum_{m_e,q,m_g} (-1)^{J_e-m_e} \begin{bmatrix} J_e & 1 & J_g \\ -m_e & q & m_g \end{bmatrix} \int_{-\infty}^{\infty} dt_0 e^{+i\omega_{eg}t_0} [\langle J_e m_e | U^{I\dagger}(t_0, -\infty) [\hat{\mu}_e^I(t_0)]_q U^I(t_0, -\infty) | J_g m_g \rangle^*]_{\text{coll}} \tag{4}$$

and

$$\mu_2 = \sum_{J_i} \mu_{2p}(J_i) e^{-\beta(E_i-E_g)} , \tag{5}$$

where

$$\mu_{2p}(J_i) = \sum_{m_e,q,m_g,m_i} (-1)^{J_e-m_e} \begin{bmatrix} J_e & 1 & J_g \\ -m_e & q & m_g \end{bmatrix}$$
$$\times \int_{-\infty}^{\infty} dt_0 e^{-i\omega_{eg}t_0} [\langle J_e m_e | U^I(\infty, t_0) [\hat{\mu}_e^I(t_0)]_q U^I(t_0, -\infty) | J_i m_i \rangle \langle J_g m_g | \hat{S} | J_i m_i \rangle^*]_{\text{coll}} . \tag{6}$$

In these equations $U^I(t, t')$ is the time evolution operator for the radiator-perturber collision and $\hat{\mu}_e^I(t)$ is the induced dipole operator in the interaction (I) picture. The subscript coll denotes an average over classical trajectory quantities, namely impact parameter and velocity.

The absorption coefficient may be parametrized as follows:

$$\alpha(\omega) = n_R \left[\frac{4\pi^2\omega}{3\hbar c} \right] (1-e^{-\beta\hbar\omega}) P(J_g) |\mu_R|^2$$
$$\times \left[\frac{bn_p/\pi}{(\Delta-Sn_p)^2+(Bn_p)^2}(1+an_p+bn_p^2) \right.$$
$$\left. - \frac{(\Delta-Sn_p)/\pi}{(\Delta-Sn_p)^2+(Bn_p)^2}(cn_p+dn_p^2) \right] , \tag{7}$$

where

$$\gamma = n_p(B+iS) , \tag{8}$$
$$\mu_1 = y_1' + iy_1'' , \tag{9}$$
$$\mu_2 = y_2' + iy_2'' , \tag{10}$$

are complex and the real parameters a, b, c, d are

$$a = (y_1' + y_2')/\mu_R , \tag{11}$$
$$b = (y_1'y_2' - y_1''y_2'')/\mu_R^2 , \tag{12}$$
$$c = (y_1'' + y_2'')/\mu_R , \tag{13}$$
$$d = (y_1'y_2'' + y_1''y_2')/\mu_R^2 . \tag{14}$$

These four parameters describe the role of the interference in the absorption process. The parameters a and c are associated with effects which occur in single collisions and which depend upon the perturber density in a linear fashion; the leading intensity interference term involves a which affects the strength of the symmetric Lorentzian contribution to $\alpha(\omega)$. The asymmetry of the profile, on the other hand, is controlled by c which determines the magnitude of the anomalous dispersion component in $\alpha(\omega)$. The parameters b and d play analogous roles to a and c but in succeeding collisions, important at higher densities. These parameters depend upon collision details averaged over two separate collisions; the averages are nonzero because in each collision there arises an induced dipole component capable of interfering with the allowed dipole and different collisions are thereby correlated. The contributions described by b and d depend on the square of the perturber density, and are small at low and moderate densities. Typically they become significant, both experimentally and theoretically, at densities of the order of 50 to 100 amagat.

III. CALCULATION

Calculations based on these equations are described in detail in Ref. [8] for the cases of HD-He and HD-Ar. The major inputs to the calculation are semiempirical expressions for the adiabatic intermolecular potentials and expressions calculated in *ab initio* fashion for the pair induced dipole moments, both of which are functions of intermolecular distance R.

The potentials used [8,10,11] are of the form

$$V(R,\theta) = \sum_{k=0}^{\infty} V^{(k)}(R)P_k(\cos\theta) , \qquad (15)$$

where θ is the angle between $\vec{R}$ and the internuclear axis of the molecule. Terms up to $k=3$ were included in the calculation. In the integrals for μ_1 and μ_2, V appears in the time evolution operator and can cause transitions among rotational states of the HD molecule. The highest J state included in the calculation is $J_{max}=4$. Calculations are performed for rotational lines $R(J)$ for $J=0$ to 2, which result from the selection rule $\Delta J=1$.

IV. INDUCED DIPOLE MOMENT

The concern of the present paper is with the induced pair dipole moment. This total induced moment has been calculated and partitioned into components of different symmetry by Borysow, Frommhold, and Meyer [12]. For an atom-diatomic molecule interaction, the spherical component q of the induced moment may be expressed as [12,13]

$$[\hat{\mu}_e(\vec{R})]_q = 4\pi \sum_{\Lambda,L} B_\Lambda^{vv'}(\Lambda,L;R) \sum_m (-1)^{1-q} \begin{bmatrix} 1 & \Lambda & L \\ -q & q-m & m \end{bmatrix} Y_{\Lambda,q-m}(\Omega)Y_{LM}(\theta,\phi) , \qquad (16)$$

where Ω is the orientation of the molecule and (θ,ϕ) specifies the orientation of $\vec{R}$. The $B_\Lambda^{vv'}$ form a coordinate system-independent classification of the components of the induced moment. For the pure rotational transitions between levels in the ground vibrational state $v=v'=0$; these superscripts will be dropped in the remainder of the paper. The Λ refers to the symmetry (i.e., tensorial rank) of the molecular charge distribution and L refers to the rank of the field tensor for the charge distribution. Borysow et al. [12] have assigned nine components of $B_\Lambda(\Lambda L)$ in (16) for the HD-X interaction. They express the R dependence of the $B_\Lambda(\Lambda L)$ as the sum of an exponential, and an R^{-n} contribution, where n is an integer. It is important to be aware of the physical significance of each component, in terms of the interaction responsible for its induction.

$B_0(01)$ arises from the isotropic overlap interaction. $B_1(10)$ has the same symmetry as the permanent moment and arises from an anisotropic overlap interaction. Five other components $B_1(12)$, $B_2(21)$, $B_3(32)$, $B_3(34)$, and

$B_4(43)$ are also induced through anisotropic overlap interactions. For all these overlap interactions the dominant R-dependent term is a short-range exponential function and the R^{-n} is also of relatively short range with n assigned the value of 7 in all cases, except $B_3(34)$, where it is 4. The component $B_2(23)$ principally arises from the long-range quadrupole interaction and n is 4. Another long-range interaction, hexadecapole moment induction, is responsible for the $B_4(45)$ component; here n is 6. The largest components, and hence the major contributors to the purely induced absorption coefficient for both HD-He and HD-Ar, are $B_0(01)$, $B_2(23)$, $B_2(21)$, and $B_3(32)$.

V. RESULTS

Tables I–III present typical results from the calculation of a, c, b, and d for HD-He and HD-Ar at temperatures of 77, 195, and 295 K. The allowed dipole element was taken as -8.3 D [14]. The contribution from each

TABLE I. Values of interference parameters for $R(1)$ of HD-He at 77 K. The experimental values are taken from Ref. [3].

Induced-dipole component	a (amagat^{-1})	c (amagat^{-1})	b (amagat^{-2})	d (amagat^{-2})
$B_0(01)$	1.17×10^{-4}	-6.08×10^{-4}	1.81×10^{-14}	-6.95×10^{-14}
$B_1(10)$	4.37×10^{-3}	3.19×10^{-5}	4.77×10^{-6}	6.72×10^{-8}
$B_1(12)$	3.07×10^{-5}	-1.10×10^{-4}	-3.33×10^{-9}	-1.90×10^{-9}
$B_2(21)$	-2.53×10^{-4}	8.61×10^{-4}	-6.97×10^{-8}	-9.23×10^{-8}
$B_2(23)$	-4.46×10^{-6}	-1.12×10^{-4}	-1.09×10^{-9}	3.21×10^{-10}
$B_3(32)$	1.05×10^{-5}	-4.66×10^{-6}	-7.91×10^{-11}	2.81×10^{-10}
$B_3(34)$	-8.90×10^{-6}	-2.26×10^{-6}	1.60×10^{-11}	8.98×10^{-12}
$B_4(43)$	-1.26×10^{-6}	-2.25×10^{-6}	-1.33×10^{-13}	1.34×10^{-12}
$B_4(45)$	-2.40×10^{-7}	1.17×10^{-6}	-3.63×10^{-13}	-5.15×10^{-14}
total	4.26×10^{-3}	5.4×10^{-5}	4.5×10^{-6} [a]	1.2×10^{-7} [a]
expt.	$6.2(7)\times10^{-3}$	$-2.6(6)\times10^{-3}$		

[a]Reference [8].

TABLE II. Values of interference parameters for $R(1)$ of HD-He at 195 K. The experimental values are taken from Ref. [3].

Induced-dipole component	a (amagat^{-1})	c (amagat^{-1})	b (amagat^{-2})	d (amagat^{-2})
$B_0(01)$	-1.42×10^{-5}	-1.65×10^{-3}	2.08×10^{-14}	-1.87×10^{-13}
$B_1(10)$	5.91×10^{-3}	7.69×10^{-6}	8.73×10^{-6}	2.35×10^{-8}
$B_1(12)$	-1.00×10^{-4}	-3.16×10^{-4}	-2.25×10^{-8}	1.59×10^{-8}
$B_2(21)$	-8.26×10^{-4}	7.08×10^{-4}	4.90×10^{-8}	-2.88×10^{-7}
$B_2(23)$	-8.04×10^{-5}	-1.06×10^{-4}	1.05×10^{-10}	3.23×10^{-9}
$B_3(32)$	-4.18×10^{-5}	1.57×10^{-5}	3.90×10^{-10}	-2.85×10^{-10}
$B_3(34)$	-1.90×10^{-5}	8.88×10^{-6}	6.76×10^{-11}	-7.97×10^{-11}
$B_4(43)$	-1.22×10^{-6}	-3.53×10^{-6}	-2.50×10^{-12}	3.02×10^{-12}
$B_4(45)$	1.90×10^{-6}	3.32×10^{-6}	-1.76×10^{-12}	3.49×10^{-12}
total	4.83×10^{-3}	-1.33×10^{-3}	6.0×10^{-6} [a]	-3.3×10^{-6} [a]
expt.	$-0.4(9) \times 10^{-3}$	$-2.18(46) \times 10^{-3}$		

[a]Reference [8].

of the various induced dipole moment components is identified.

The following discussion considers each parameter in turn and gives a general description of its origin.

A. Intensity parameter a

As is evident from (4), (5), (9), (10), and (11), the total a is simply the result of the sum of the contributions from the individual induced dipole components.

The largest contribution in all cases comes from $B_1(10)$. This result is not surprising and is, in fact, expected. Since $B_1(10)$ has the same symmetry as the allowed dipole moment, it may interfere, that is, produce a nonzero cross term with the allowed moment, even for a purely isotropic HD-X interaction. It is this interaction scheme which formed the basis of the pioneering theory of the interference effect by Tipping, Poll, and Herman [4–6].

Other induced dipole components cannot contribute directly to the interference effect. They can be involved only through the action of an anisotropic intermolecular interaction which mixes molecular rotational levels. The tables show that the contribution of all of these other dipole components is at least one order of magnitude smaller than, and most often of the opposite sign to, the $B_1(10)$ component. Their net effect is usually to reduce the magnitude of a by an amount up to 70%.

An exception to these statements is $R(0)$ for HD-Ar at 295 K where the $B_2(21)$ contribution is large. Also for $R(2)$ at 77 and 195 K for both HD-He and HD-Ar, the overall effect of the inclusion of rotational level mixing is to increase the value of a over that calculated when only an isotropic interaction is considered [8].

In general, the contribution of the quadrupole induced components $B_2(23)$ is larger for HD-Ar than for HD-He. This result might be expected because of the much larger $B_2(23)$ component in the induced moment itself for HD-Ar, as a result of the larger polarizability of argon compared to that of helium.

B. Asymmetry parameter c

As in the case of a, the net value for c is the simple sum of the contributions from each induced dipole com-

TABLE III. Values of interference parameters for $R(1)$ of HD-Ar at 295 K. The experimental value is taken from Ref. [2].

Induced-dipole component	a (amagat^{-1})	c (amagat^{-1})	b (amagat^{-2})	d (amagat^{-2})
$B_0(01)$	-2.31×10^{-4}	-1.82×10^{-3}	-4.03×10^{-14}	-1.23×10^{-13}
$B_1(01)$	8.35×10^{-3}	-2.27×10^{-5}	1.74×10^{-5}	-8.56×10^{-8}
$B_1(12)$	-2.35×10^{-4}	-2.84×10^{-4}	-5.69×10^{-9}	3.43×10^{-8}
$B_2(21)$	-3.59×10^{-3}	2.44×10^{-3}	1.72×10^{-6}	-4.37×10^{-6}
$B_2(23)$	-9.53×10^{-4}	-3.85×10^{-4}	2.18×10^{-7}	1.08×10^{-7}
$B_3(32)$	-1.29×10^{-4}	-1.10×10^{-4}	1.07×10^{-9}	7.10×10^{-9}
$B_3(34)$	-9.65×10^{-5}	9.69×10^{-5}	1.56×10^{-10}	-4.67×10^{-9}
$B_4(43)$	3.79×10^{-7}	-7.49×10^{-6}	-1.52×10^{-11}	1.36×10^{-12}
$B_4(45)$	2.60×10^{-5}	3.47×10^{-6}	1.66×10^{-10}	4.78×10^{-11}
total	3.41×10^{-3}	-8.88×10^{-5}	3.4×10^{-6} [a]	-1.2×10^{-6} [a]
expt.	$1.8(3) \times 10^{-3}$			

[a]Reference [8].

ponent. Unlike a, however, for this profile asymmetry parameter, in all cases the $B_1(10)$ contribution is completely negligible. The purely isotropic molecular interaction is thus unimportant. The major contributions come from $B_0(01)$, $B_2(21)$, or $B_2(23)$, the largest components of the dipole moment and those which have the largest effect on the purely induced absorption. There is, moreover, considerable cancellation among the contributions from these components, with the net value of c differing greatly in magnitude and even in sign from that of the largest single contribution.

The unimportance of the purely isotropic interaction involving $B_1(10)$ is, perhaps, surprising, given that it is the major contributor to the intensity interference effect and that asymmetric profiles, particularly for the vibration-rotation transitions, have come to be associated with collisional interference in the HD spectrum [15]. However, the concern here is with details of the profile, not the total intensity. An anisotropic interaction is necessary for collision broadening to occur. The isotropic intermolecular potential and $B_1(10)$ component alone would lead to symmetric extremely sharp lines [16]. Thus the total intensity interference effect, on the one hand, and line broadening and profile asymmetry, on the other, are seen to be sensitive to different details of the HD-X interaction.

C. Intensity parameter b

Assessment of the contributions of the dipole moment components to the parameters b and d is a more difficult task. They describe terms in the absorption coefficient which depend on the perturber density squared and which arise from events occurring in two separate collisions. They thus depend upon the product of μ_1 and μ_2 [Eqs. (4) and (5)] and all components of the induced dipole appear in the integrands, averaged over a collision, for both μ_1 and μ_2. In the calculation there are then 81 contributions to b and d, involving various products of pairs of components of the induced moment. To make an assessment of the relative importance of the various components, the only contributions of b and d reported in the tables are those where a given single induced dipole component is taken to act in both collisions. For b, an intensity parameter, the $B_1(10)$ component is again the most important and the effect of $B_2(21)$, the next most important, is an order of magnitude, or more, smaller. For HD-Ar, $B_2(23)$ is also non-negligible. If one compares these values with the total value of b given in Ref. [8], it is seen that in all cases the total b is smaller, by up to an order magnitude, than the contribution of $B_1(10)$. The terms containing products of different components of the induced moment are therefore significant. The mixing process again has the effect of reducing the intensity interference from that obtained with a purely isotropic interaction. It is of interest to note that the contributions to b listed in the tables are not all positive. This result emphasizes the fact that the μ_1 and μ_2 are not equal for a given interaction.

D. Asymmetry parameter d

For the asymmetry parameter d, $B_2(21)$ always gives the most important term, with $B_1(10)$ and $B_1(12)$ ranking next for HD-He and with $B_2(23)$ second most important for HD-Ar. Components of similar magnitude often have different sign and cancellation among various contributions is substantial especially when all possible cross terms are considered. Comparison with the full values of d tabulated in Ref. [8] shows that they are smaller or larger than the pure contribution of $B_2(21)$ by up to an order of magnitude, and occasionally differ in sign.

VI. DISCUSSION

The theoretical formalism providing the basis of these calculations was developed for the express purpose of giving a more general description of the interference phenomenon than was previously available. Specifically it represents an improvement in that it allows for effects due to m and J mixing [7]. The calculations performed with it in Ref. [8] show that contributions of inelastic rotational collisions to the interference and line broadening mechanisms are essential. In a theory in which the intermolecular potential is taken to be the adiabatic potential averaged over m states, the m- and J-changing collisions are ignored and the only contribution to the interference effect comes from the $B_1(10)$ component of the induced dipole [8]. Contributions from other dipole components thereby correlate strongly with m- and J-changing processes, or, to state the situation in other terms, with the importance of the anisotropic part of the HD-atom potential. The greater the m- and J-changing cross sections, the greater are the contributions from the other components. A good measure of m and J processes is the pressure broadening coefficient. Two approximate rules follow from these considerations. First, the broader the line, the more significant should be the contributions from components other than $B_1(10)$. This tendency can be seen by comparing the results for HD-He and HD-Ar. In Ref. [8] it is shown that the pressure broadening coefficient for HD-Ar is two to three times larger than that for HD-He. Inspection of Tables I, II, and III shows that these other dipole components are more important for HD-Ar than for HD-He. Second, the higher the temperature, the more important should be contributions from the other components. Both the broadening coefficient [8] and the contributions from the other components to the interference parameters increase with temperature (Tables I and II). These comparisons also demonstrate that the calculation is sensitive to the potential model.

The uncertainty given for the experimental values in the tables represents one standard deviation. For an extensive comparison of calculation and experiment the reader is referred to Ref. [8]. Some general statements, however, will be made here. The semiclassical calculation overestimates the inelastic transition rates at low collision energies. The calculated results for $R(0)$ at low temperature are therefore the least reliable with the better results expected to be for the $R(J)$ with $J > 0$ at

high temperatures. The interference parameters were found to be quite sensitive to the region around the turning point of the trajectory (at the distance of closest approach) where the induced dipole has the largest magnitude. This is the region where a classical trajectory description is the least valid. A fully quantum-mechanical theory may be necessary to produce good quantitative agreement between calculation and experiment.

If inelastic collisions are neglected, the interference parameters a and b show little dependence on J and the asymmetry parameters turn out to be extremely small. The present results generally are of the correct order of magnitude and show features not contained in earlier approaches. They clearly demonstrate that inclusion of inelastic collisions is necessary and that contributions of components other than $B_1(10)$ to the interference process are possible.

Highly quantitative statements about the precise role of the various dipole components cannot be made. Nevertheless their relative importance can be assessed and some general conclusions may be drawn.

The most important term for both single and succeeding collision contributions to the intensity interference is $B_1(10)$, the anisotropic overlap-induced dipole component which has the same symmetry as the permanent moment. It does not, however, contribute to the profile asymmetry. The most important component affecting asymmetry is $B_2(21)$, which is the largest anisotropic component in the pair induced dipole.

Rotational level mixing by the anisotropic intermolecular potential is important, affecting significantly the magnitude of the intensity interference and making possible line broadening and asymmetry. Previously it had been thought that an important effect of mixing would be the making possible of a contribution to the interference process from the very large isotropic overlap-induced component of the pair dipole moment $B_0(01)$. It is seen, however, that other components, $B_2(21)$ and $B_2(23)$, can have a much greater effect on the interference parameters. The fact that mixing almost always lowers the magnitude of the intensity interference was also not predicted.

The component $B_3(32)$, which has about the same magnitude as $B_2(21)$ in the induced dipole expression, is quite unimportant to the interference parameters. This is because it can only enter the interference mechanism through a mixing process which has a low probability. Similarly the $B_4(43)$ and $B_4(45)$ components are weak, require an improbable mixing process to play a role in the interference, and are completely negligible.

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