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INTERPRETATION OF EXPERIMENTAL RESULTS ON THE RELAXATION OF OPTICALLY PUMPED Rb IN COLLISIONS WITH Kr ATOMS

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A theoretical analysis of experimental results on the relaxation induced by Rb-Kr collisions reveals the existence of two very different correlation times. The shorter, of the order of 10^{-12} sec, is a kinetic collision time; the longer, 10^{-8} sec, reflects the presence of metastable states associated with resonances in Rb-Kr scattering, or of actual Rb-Kr bound states due to three-body collisions.

Experimental results on the relaxation of observables $\langle S_z\rangle$ and $\langle\vec{S}\cdot\vec{I}\rangle$ of optically pumped Rb in collisions with Kr gas atoms have been reported recently.[1] It has been shown that once the effect of wall relaxation has been removed as stated in paragraph 5 of PR1, all time constants which appear in the measurements are indeed due to collisions in the gas phase. Experimentally, two time constants τ_e and τ_n appear in the relaxation for $\langle S_z\rangle$, and one, τ_H, for $\langle\vec{S}\cdot\vec{I}\rangle$. A very unexpected result, clearly seen in Fig. 1 of PR1, is the very strong field dependence of $1/\tau_e$ and $1/\tau_n$, between 0 and 200 G; $1/\tau_H$, on the other hand, is field independent. Our purpose here is to show that, as announced in PR1, the observed behavior can be explained by assuming the existence of two uncorrelated interactions I and II whose physical nature is not necessarily different, but with very different correlation times τ_{c1} (very long, ~10^{-8} sec) and τ_{c2} (10^{-12} sec).

For a random weak interaction of the magnetic type $\gamma_S\vec{S}\cdot\vec{H}(t)$, theory[2] indeed predicts that two time constants τ_e, τ_n appear on $\langle S_z\rangle$, and one, τ_H, on $\langle\vec{S}\cdot\vec{I}\rangle$; furthermore, every relaxation rate can be expressed in terms of the Fourier transform of the correlation function of the perturbation $j(\omega)=(1+\omega^2\tau_c^2)^{-1}$ at frequencies $\omega=\omega_F$, Zeeman frequency, and $\omega=\Delta W$, hfs interval. For interaction II, if τ_{c2} is so short that $\tau_{c2}\omega_F\ll 1$ in the field explored, and $\tau_{c2}\Delta W\ll 1$, then the relations $j_2(\omega_F)=j_2(\Delta W)=1$ hold; this implies $\tau_{c2}\approx 10^{-12}$ sec. For the very long interaction I, on the other hand, the field dependence of the relaxation rates $1/\tau_n$ and $1/\tau_e$ suggests $\tau_{c1}\Delta W\gg 1$ and $j_1(\Delta W)\approx 0$. Taking these relations into account, one obtains[2]

$$1/\tau_H=C_2;$$

$$\frac{1}{\tau_e}=\frac{C_1}{(2I+1)^2}j_1(\omega_F)+C_2;$$

$$\frac{1}{\tau_n}=\frac{C_1}{(2I+1)^2}j_1(\omega_F)+\frac{2C_2}{(2I+1)^2},$$

with $C_1=\frac{2}{3}\gamma_S^2\langle|H(t)|^2\rangle\tau_{c1}$ and a similar expression for C_2. These formulas describe all observed facts in the range of fields explored. In high fields (about 200 G), $j_1(\omega_F)\ll 1$, interaction II is dominant. One has $\tau_H=\tau_e{}^*=2\tau_n{}^*/$

$(2I+1)^2$ ($=\frac{1}{8}\tau_n^*$, for ^{87}Rb). This exactly what is observed (see paragraphs 5 and 7 of PR1). τ_n^* and τ_e^* are then the decay constants (due to interaction II) of the (exponential) relaxations of $\langle I_z\rangle$ and $\langle Q_e\rangle$.[2] This leads to the following values for the disorientation cross sections[3] of observables $\langle \vec{S}\cdot\vec{I}\rangle$, $\langle Q_e\rangle$, and $\langle I_z\rangle$ of ^{87}Rb: $\sigma_H=\sigma_e=8\sigma_n=(27\pm3)\times10^{-21}$ cm^2 (this is also the disorientation cross section σ for a Rb isotope which would have zero nuclear spin). Interaction II and the relaxation at 200 G behave exactly as expected for collisions in the gas phase: $\tau_{c2}\approx10^{-12}$ sec is of the proper order for a "collision time." Moreover, the relaxation rate (PR1, paragraph 4) is proportional to p, i.e., to the collision frequency. It seems likely that interaction II is the spin-orbit $\vec{S}\cdot\vec{N}$ interaction proposed by Bernheim.[4]

In low fields, on the other hand, the observed H_0 dependence of τ_e and τ_n (PR1, paragraph 3) is due to $j_1(\omega_F)$. τ_{c1} can be deduced from the width ΔH_0: $\tau_{c1}=(2I+1)/\gamma_S\Delta H_0$, and is of the order of 10^{-8} sec. Such a long correlation time is most unexpected: It is about 10^4 times longer than a "collision time" (10^{-12} sec) in the gas phase. As stated in PR1, paragraph 6, τ_{c1} gets shorter when p increases (23×10^{-8} sec at 0.2 Torr and 18×10^{-8} sec at 2 Torr); it is close to, but somewhat shorter than, the time of flight τ_v between two successive collisions of a Rb atom on Kr ($\tau_v\approx133p^{-1}$ nsec for p in Torr). As seen in Fig. 1 of PR1, interaction I is dominant in low fields, in the pressure range explored, and C_1 has clearly a complicated, and unexpected, behavior with p (PR1, paragraph 6).

It seems that a likely explanation of those facts goes along the following lines. A Rb-Kr collision can be described by the motion of a single particle in a potential $V_{\rm eff}^N(r)=V(r)+N(N+1)\hbar^2/2mr^2$, $V(r)$ being the potential energy of the two atoms at rest at distance r,[5] $N\hbar$ their relative orbital angular momentum, and m the reduced mass. At room temperature, the shape of $V_{\rm eff}^N(r)$ is as shown in Fig. 1: There is a potential well protected by a potential barrier of centrifugal origin. Two kinds of processes can produce Rb-Kr complexes having a long lifetime. First, whenever the relative kinetic energy of the two colliding atoms coincides with one of the quantized levels E_{jN} of the relative particle inside the well, a metastable state of lifetime τ_{jN} has a probability of being produced, proportional to τ_{jN}^{-1}. Second, an actual Rb-Kr bound state ($E_{jN}<0$) can also be produced during a three-body collision involving one Rb and two Kr atoms.[6] In both cases, the Rb-Kr complex is very likely destroyed in its next collision with another Kr atom, so that its effective lifetime $\tau_{\rm eff}$ depends on p. In such a complex, the disorienting $\vec{S}\cdot\vec{N}$ interaction acts on Rb during the time $\tau_{\rm eff}$. One can thus understand the existence of a long correlation time τ_{c1} of the order of $\tau_{\rm eff}$ and becoming shorter when p is increased, in agreement with the results of paragraph 6 of PR1. A similar behavior is expected when a foreign gas, like He, is added: Collisions with He destroy the Kr-Rb pair and reduce τ_{c1}. Figure 2 shows the experimental results obtained for

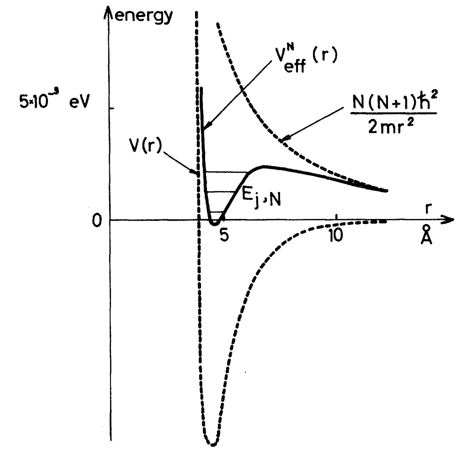

FIG. 1. One-dimensional potential $V_{\rm eff}^N(r)$ used to describe the motion of the relative particle during a Rb-Kr collision occurring with the value $N\hbar$ of the relative angular momentum. The Lennard-Jones potential $V(r)$ is taken from sources indicated in Ref. 5.

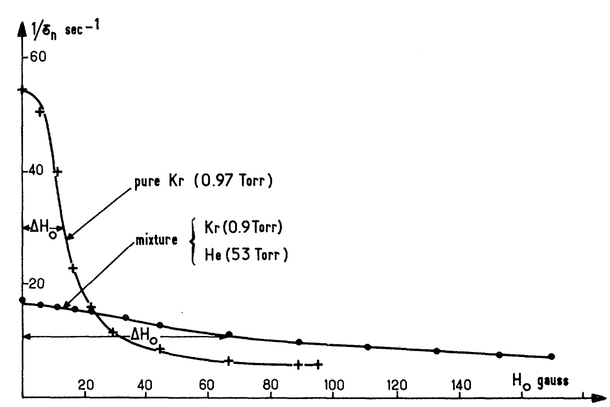

FIG. 2. Relaxation rate of $\langle I_z\rangle$ measured on ^{87}Rb in pure Kr (pressure 0.97 Torr) and in a mixture of Kr (partial pressure 0.9 Torr) and He (53 Torr) versus the static magnetic field H_0.

the field dependence of $1/\tau_n$ in pure Kr (0.97 Torr) and in a mixture of Kr (0.9 Torr) and He (53 Torr). At the same Kr pressure the relaxation rate in low fields is decreased in the presence of He. This comes indeed from the shortening of τ_{c1} as is also revealed by the broadening of the curve giving the field dependence of $1/\tau_n$. As expected, in high fields relaxation rates are the same: Direct measurements in pure He had previously shown that Rb-He collisions have a negligible effect at 53 Torr.

A detailed analysis, to be published elsewhere, shows that the above picture leads to a satisfactory quantitative description of the main features of interaction I. The metastable states of the alkali-metal–rare-gas pair have not been observed directly in scattering experiments; such an observation seems unlikely because the unrelated resonances are so narrow ($\hbar\tau_{jN}^{-1}/kT\approx10^{-6}$). On the other hand, the relaxation of alkali metals is obviously very sensitive to their existence; their probability of being formed is indeed small, but the time during which the disorienting interaction lasts and its correlation time are approximately 10^4 times longer than in ordinary collisions. The same remark holds for actual bound states.

We have observed a very similar behavior of the relaxation of Rb in Xe and Ar.

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[1]M. Aymar, M. A. Bouchiat, and J. Brossel, Phys. Letters 24A, 753 (1967), hereafter referred to as PR1.

[2]M. A. Bouchiat, J. Phys. Radium 24, 379, 611 (1963); M. A. Bouchiat and J. Brossel, Phys. Rev. 147, 41 (1966).

[3]Any observable Q_i relaxing exponentially with time τ_i is associated with a disorientation cross section σ_i given by $1/\tau_i=N_0\sigma_i\bar{v}_{\rm rel.}p/p_0$; N_0 is the number of atoms per cc at pressure p_0; $\bar{v}_{\rm rel.}$ is the relative velocity.

[4]R. A. Bernheim, J. Chem. Phys. 36, 135 (1962).

[5]A. Dalgarno and A. E. Kingston, Proc. Phys. Soc. (London) 73, 455 (1959); Fr. von Busch, H. J. Strunck, and Ch. Schlier, Z. Physik 199, 518 (1967).

[6]We are indebted to Dr. Bender for stressing the importance of three-body collisions.

GENERATION OF A PHASE-MATCHED OPTICAL THIRD HARMONIC BY INTRODUCTION OF ANOMALOUS DISPERSION INTO A LIQUID MEDIUM

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The use of anomalous dispersion in producing phase matching in nonlinear optical processes has been recognized in several references in the nonlinear-optics literature,[1-4] and the anomalous dispersion associated with the strong infrared resonance inherent in quartz has been employed in producing a far-infrared difference frequency.[5] The present Letter provides the first experimental evidence that phase matching may be achieved in a nonlinear optical process by introduction of anomalous dispersion into a normally unmatched medium.

The principle is indicated schematically in Fig. 1. Curve a shows the normal dispersion of a medium and the typical index mismatch, $\Delta n=n_h-n_f$, between two frequencies ω_h and ω_f. In the harmonic generation process here under consideration, these are the harmonic

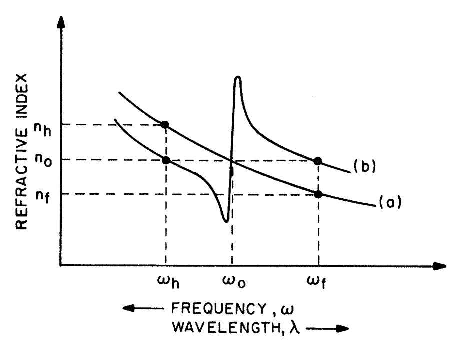

FIG. 1. Principle of phase matching by anomalous dispersion. Curve a shows a normally dispersive medium with $n_h>n_f$ at the frequencies ω_h and ω_f. Curve b, the same medium after the introduction of anomalous dispersion centered at ω_0, resulting in identical indices n_0 at the two frequencies.