Research paper

Development of NQR explosive detection technique for transportation security

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Abstract

Nuclear Quadrupole Resonance (NQR) technique is expected as a promising method for explosive detection, since some major explosives are known to possess the substance−specific NQR frequency. We have developed a prototype of NQR explosive detection system and examined the applicability of NQR technique to explosive detection for transportation, in particular, aviation security. In this study, we examined two types of surface coils, nonlinear spiral and gradiometer−type, and several large volumetric solenoids as sensors for detection of explosives hidden in shoe and baggage, respectively. The limit distance for detection of 500 g of $NaNO_2$ was *ca.* 8.5 cm above the surface of gradiometer coil without electromagnetic shield. The gradiometer coil also clearly detected 300 g of hexahydro−1, 3, 5−trinitro−1, 3, 5−triazine (RDX) located 3.5 cm above the coil−surface without the shield. A blind test of NQR as well as X−ray baggage screening tests were conducted with 50 baggage−samples containing RDX or non−explosive materials at the security−check gate in the airport. The resulting false positive rate of NQR screening with large solenoid was about one−tenth of that of the conventional X−ray inspection. It indicates that the security−check by NQR explosive detection technique is useful for the transportation security.

Keywords : RDX, Explosive detection, NQR, Aviation security

1. Introduction

Transmitting X−ray imaging is currently used for shoe or baggage screening at the airport to detect explosive compounds. X−ray screening can distinguish the shape and the X−ray transmittance of objects in baggage, but its false positive alarm rate is significantly high. This is because there are varieties of food products and substances which have the similar X−ray transmittance to organic explosives such as transformable plastic explosives. Meanwhile, Nuclear Quadrupole Resonance (NQR) technique can be used for detection with the identification of the substance including explosives, since the NQR frequencies intrinsically depend on their crystal structure[1)−8)]. Many of explosives contain nitrogen atoms, and a nucleus of ^{14}N (natural abundance of 99.63 %) has a nuclear spin of quantum number of I=1. For the nucleus with I=1, three generally non−degenerated quadrupole energy levels are formed by the electrostatic interaction between the nuclear quadrupole moment, eQ, and the second derivative of the electric potential (the electric field gradient, eq) of the surrounding electron cloud. Three NQR frequencies among the three energy levels are given as $\nu_+ > \nu_- > \nu_0$:

$$\nu_+ = \frac{e^2Qq}{4h}(3+\eta) \qquad (1)$$

$$\nu_- = \frac{e^2Qq}{4h}(3-\eta) \qquad (2)$$

$$\nu_0 = \nu_+ - \nu_- = \frac{e^2Qq}{2h}\eta \qquad (3)$$

Where e^2Qq/h is the quadrupole coupling constant in frequency units and η is the asymmetry parameter of the electric field gradient (EFG) tensor. The quadrupole moment eQ is a nuclear physics parameter describing the charge distribution in the nucleus and is a constant for a given nucleus. The EFG tensor, however, largely depends on chemical environments of the atom in question. Not only the distribution of valence electrons around the nucleus but also that of the external charge such as surrounding ions or molecules, contribute to the EFG. Therefore, the NQR frequencies are unique quantities depending on the crystal structure of substances. This fact allows the identification of a specific substance by NQR technique, though the NQR frequency varies slightly according to the temperature in general.

A multi−pulse method is thought to be suite for the purpose of shoe and baggage screening. A strong off−resonance comb (SORC) pulse sequence is described as $(t-\phi-t)_N$, where ϕ is the flip angle defined as $\phi = \gamma H_1 t_w$, γ is a gyromagnetic ratio, which is nucleus−specific value, H_1 is RF magnetic field amplitude and t_w is pulse width, and $2t$ is the time interval between the pulses. The RF pulse repetition time 2τ described below is defined as $2\tau = 2t + t_w$. In this method, a continuous steady−state signal can be produced when the RF pulse repetition time 2τ is less than or equal to the spin−spin relaxation time T_2, and the signal generation in approximately every T_2^* seconds allows a very efficient signal averaging resulting in remarkable shortening of inspection time[9]. Here, the inverse line−width parameter T_2^* is generally less than T_2 and is typically~1 ms.

2. Experimental

As test substances, 414 g of hexamethylenetetramine (HMT), $(CH_2)_6N_4$ and 500 g of sodium nitrite, $NaNO_2$, both of which were immersed in liquid paraffin to prevent piezoelectric noise signal, and composites of 300 g and 500 g of hexahydro−1, 3, 5−trinitro−1, 3, 5−triazine (RDX) which were deactivated with wax[10] were used. ^{14}N NQR frequencies are 3306.2 kHz at 299.6 K for HMT [11], ν_+ = 4640 kHz and ν_- = 3603.7 kHz at 295 K for $NaNO_2$[12], ν_+ = 5240, 5192, 5047 kHz and ν_- = 3458, 3410, 3359 kHz at 298 K for RDX [13].

For a shoe screening, two types of surface coils were made. One was a six turns spiral coil of 24 cm in diameter shaped according to nonlinear spiral with the radius $a = \rho\ln(1+k\theta)$, where ρ = 5.1 cm, k = 0.25 and θ denotes the angle in radian[14], consisting of a copper wire 2 mm in diameter. The other surface coil, gradiometer coil[15], was composed of parallel−connected two spiral coils wound seven turns in opposite direction each in a diameter of 18 cm. RG 213 co−axial cables removed the outer shield were used to make the spiral coils of the gradiometer. The inductance L and quality factor Q of these surface coils were L = 11 μH, Q = 100 at 4.6 MHz, and L = 4 μH, Q = 120 at 4.64 MHz, respectively.

For a baggage screening, two types of large solenoids were made. One was by winding the outer−shield−removed co−axial cable three turns on a frame of 45 cm (height) × 45 cm (width) × 34 cm (depth) size, and the other was by winding a thin copper−plate band (0.2 mm thick 18 mm in width) two turns on a frame of 30 cm (height) × 60 cm (width) × 45 cm (depth) size made by 5 mm thick acryl plates. The latter solenoid was comparable in size to the limit of carry−on baggage. The inductance L and quality factor Q of these solenoids were L = 9 μH, Q = 100 at 3.30 MHz, and L = 4 μH, Q = 130 at 5.19 MHz, respectively.

All coils mentioned above were used for both as a transmitter and a receiver coil in our NQR system. For the surface coils, distance and positional dependence of NQR signal amplitude were measured. Baggage screening test using the deactivated RDX was conducted with the acryl−frame solenoid.

The measurements were conducted with high−speed multi−pulse method using a SORC pulse sequence mentioned above. A PC−controlled pulse Fourier−transform NQR spectrometer schematically shown in Fig. 1 was con-

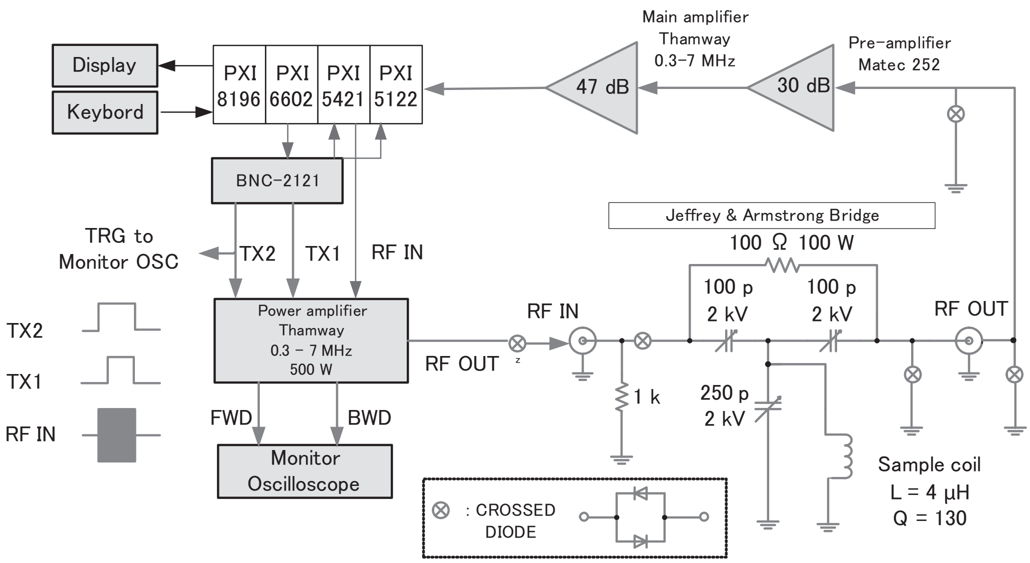

Fig. 1 Block diagram of a PC−controlled pulse Fourier−transform NQR spectrometer

structed. RF pulses were generated by a Compact PCI (PXI) module of direct digital synthesis (DDS), National Instruments PXI−5421 arbitrary waveform generator. The RF pulses were amplified by a RF power amplifier (Thamway, 0.3−7 MHz, 500 W) and transferred to the coil. During RF pulse irradiation, NQR signals emitted from samples were received by the same coil and amplified by pre−amplifier (Matec 252, 30 dB) and Main amplifier (Thamway, 0.3−7 MHz, 47 dB). As a duplexer, *i.e.* a transmit−receive switch, we used Jeffrey and Armstrong asymmetrical RF −bridge[16], which ensured a proper impedance matching when it was balanced. In the early−stage experiments, a conventional phase−sensitive detection using a double balanced mixer (DBM), a signal amplification with a low−pass filter (Tektronix AM502, 20 dB, 10 kHz), and an accumulation using a signal averager (Electronica ELK−5150AVE) were employed without fast Fourier transform (FFT). After improvement of the system, signals received by the coil were acquired directly to onboard memory of a high−speed digitizer, PXI−5122. The acquired data were transferred to PC memory and all the signal processing mentioned above, quadrature detection, low−pass filtering, accumulation, and a complex FFT were executed by a software program written in a graphical programming language, LabVIEW 8.5. All PXI modules and RF power amplifier were synchronized with 40 MHz clock generated by a counter/timers module, PXI−6602, using the same program.

3 Results and Discussion

3.1 Remote detection by surface coil

To detect explosives hidden in shoes without taking them off, the remote detection using surface coil is preferable. In the first place, we tested a nonlinear spiral coil which can improve the uniformity of RF field[14]. The NQR system without software process and FFT was used for this experiment. The amplitude of ^{14}N NQR signal ν_+ (4640 kHz at room temperature, RT) from 500 g of $NaNO_2$ was measured at RT as a function of the distance between the sample and the coil−surface. The probe part, the coil and a test sample, was put in the electromagnetic shield box. A SORC pulse sequence with the pulse amplitude of V_{pp} = 250 V for 50 Ω load, the pulse width of t_w = 200 μs, and the repetition time of 2τ = 2 ms was employed. The number of accumulation was N = 40000. The time required for the accumulation was *ca*. 8 minutes. The distance dependence of NQR signal amplitude is shown in Fig. 2 (●).

The signal amplitude was evaluated as a signal to noise ratio (S/N). The limit of distance for detection of 500 g of $NaNO_2$ was estimated to be *ca*. 18.5 cm above the surface of the coil.

With the other surface coil composed of parallel−connected two spiral coils, the gradiometer coil, the similar measurements were made. The pulse conditions were V_{pp} = 200 V for 50 Ω load, t_w = 100 μs, 2τ = 2 ms, and N = 40000. The ratios of NQR signal amplitude to noise are also shown in Fig. 2 (▲) as a function of the distance between the sample and the coil surface. The limit of distance for detection of 500 g of $NaNO_2$ was estimated to be *ca*. 15 cm

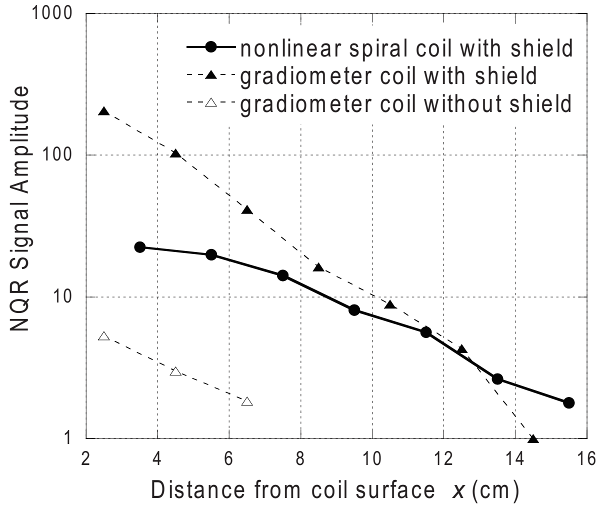

Fig. 2 Remote detection of NQR signals emitted from 500 g of $NaNO_2$ with a nonlinear spiral coil (●), with a gradiometer coil (▲) under an electromagnetically shielded circumstance, and with the gradiometer coil out of the shield (△). These measurements were conducted with the system of the earlier setup with accumulation N = 40000. The conditions of SORC pulse sequence were V_{pp} = 250 V, t_w = 200 μs, 2τ = 2 ms, and V_{pp} = 200 V, t_w = 100 μs, 2τ = 2 ms, respectively.

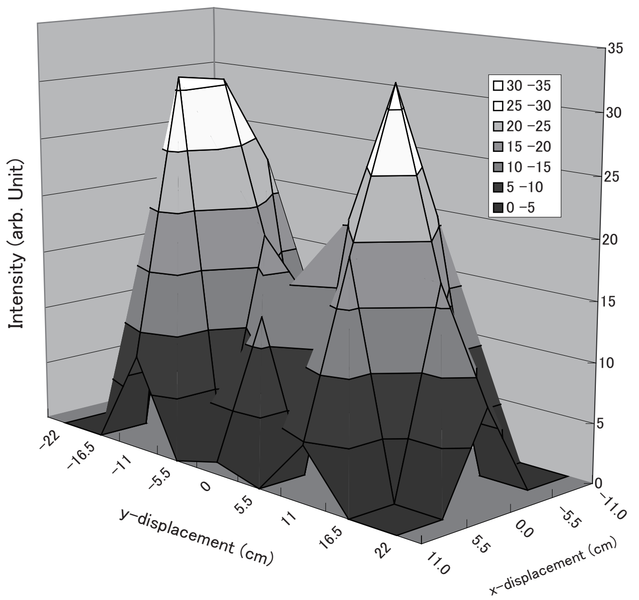

Fig. 3 Positional dependence of the NQR signal amplitude from 500 g of $NaNO_2$ on the surface of the gradiometer coil. The measurements conditions by SORC pulse sequence were V_{pp} = 200 V, t_w = 100 μs, 2τ = 2 ms, N = 40000.

above the surface of one of the component coils. The gradiometer coil has an advantage of canceling the voltage induced by the environmental electromagnetic field and so the detection of the NQR signal of 500 g of $NaNO_2$ without electromagnetic shield was tested. The distance dependence of NQR signal is also shown in Fig. 2 (△). Without the electromagnetic shield, the limit distance for detection of 500 g of $NaNO_2$ was estimated to be *ca*. 8.5 cm above the surface of one of the component coils. The positional dependence of the signal amplitude on the surface of the gradiometer coil was also measured in the shied. This is shown in Fig. 3.

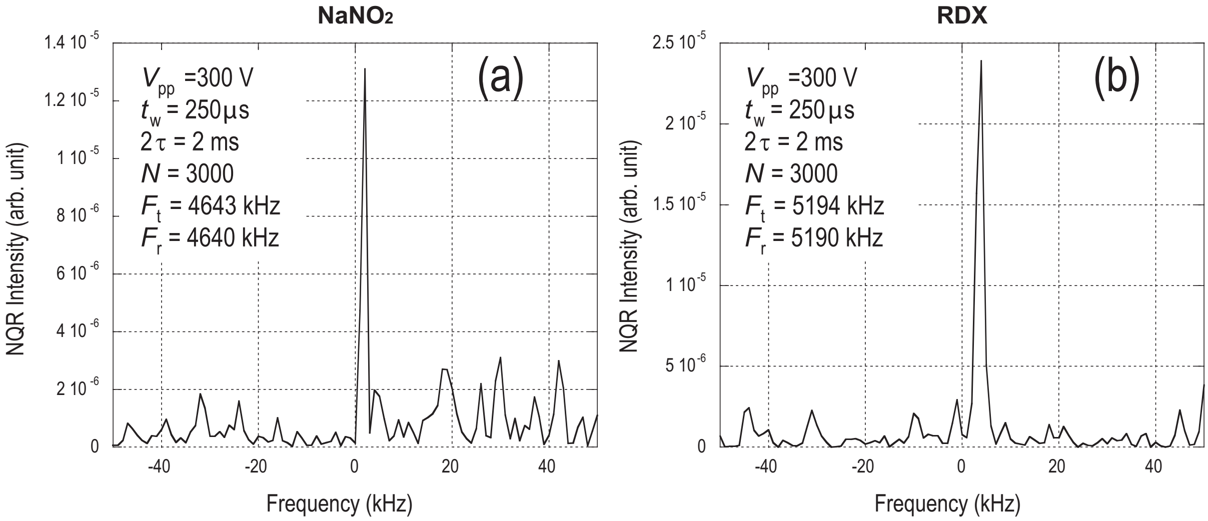

Fig. 4 (a) The signal ν_+ (4640 kHz at RT) obtained from 500 g of $NaNO_2$ located 15.5 cm above the nonlinear spiral coil. (b) The one of ν_+ lines (5192 kHz at RT) obtained from 300 g of RDX located 8.5 cm above the nonlinear spiral coil. In the insert legend, F_t and F_r denote the transmitter and reference frequencies, respectively. The abscissa is the offset from the reference frequency.

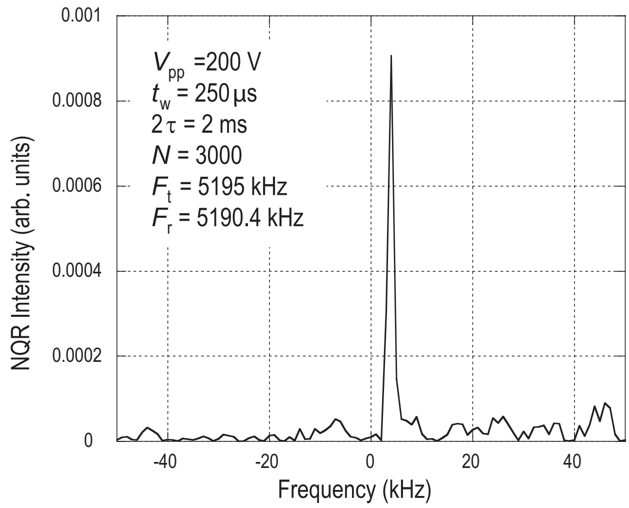

Fig. 5 The one of ν_+ lines (5192 kHz at RT) obtained from 300 g of RDX located 3.5 cm above the gradiometer coil by the FFT method under unshielded circumstance. The abscissa is the offset from the reference frequency.

Improvement of S/N was achieved by a complex FFT of the signal after digital phase-sensitive quadrature detection on the software program. By introducing FFT technique, the better result could be obtained with the smaller accumulation number of N = 3000. The signals obtained by the FFT method under shielded circumstance are shown in Fig. 4 ; (a) the ν_+ line of 500 g of $NaNO_2$ and (b) one of the ν_+ lines (5192 kHz at RT) of 300 g of RDX located 15.5 cm and 8.5 cm above the nonlinear spiral coil, respectively.

By using the FFT method, the gradiometer coil could detect the ν_+ line of 300 g of RDX located 3.5 cm above the coil-surface with S/N = *ca*. 15 under unshielded circumstance as shown in Fig. 5.

These results confirmed the feasibility of shoe screening without taking them off by using NQR technique.

3.2 Explosive detection by large volumetric solenoid

For bulk detection of explosives in carry-on baggage, the detection using a large solenoid is preferable. A large solenoid made by winding RG213 co-axial cable removed its outer shield three turns on a frame of 45 cm × 45 cm × 34 cm size was successfully used to detect the ^{14}N NQR signal (3306 kHz at RT) from 414 g of HMT as well as the ν_+ from 500 g of $NaNO_2$. The NQR signals were obtained with S/N = *ca*. 5 by use of the signal averager without FFT from the $NaNO_2$ included in a baggage and the HMT for the accumulation of 8192 and 2048, respectively. The conditions of SORC pulse sequence were as follows : the pulse amplitude of V_{pp} = 200 V for 50 Ω load, the pulse width of t_w = 200 μs, and the repetition time of 2τ = 2 ms.

Based on the above results, a solenoid for security check of carry-on baggage was also made by winding a thin copper-plate band (0.2 mm thick 18 mm in width) two turns on a frame of 30 cm × 60 cm × 45 cm size (carry-on baggage limitation) made by 5 mm thick acryl plates. The picture of the coil with RDX and the obtained FFT spectrum of NQR signal of RDX under shielded circumstances are shown in Fig. 6.

The FFT were conducted for the signal in the last half period of the repetition time 2τ in order to remove the influence of the noise due to the coil ringing. With this solenoid and the FFT method, the ability of detecting RDX hidden in a baggage was tested in the laboratory as well as in the airport. At first, influence of metallic objects was tested in the laboratory, because they had a possibility to interfere with the RF propagation between the coil and samples. The picture of baggage sample, including RDX and metallic objects, and the NQR signal from it are shown in Fig. 7.

It was shown that the NQR signal of RDX could be well detected, even metallic objects were contained in baggage. The probe, however, had to be readjusted for impedance matching. This is thought to be caused by the change of the magnetic permeability in the coil by inserting the metallic objects.

At the security check gate in the airport, a blind test of

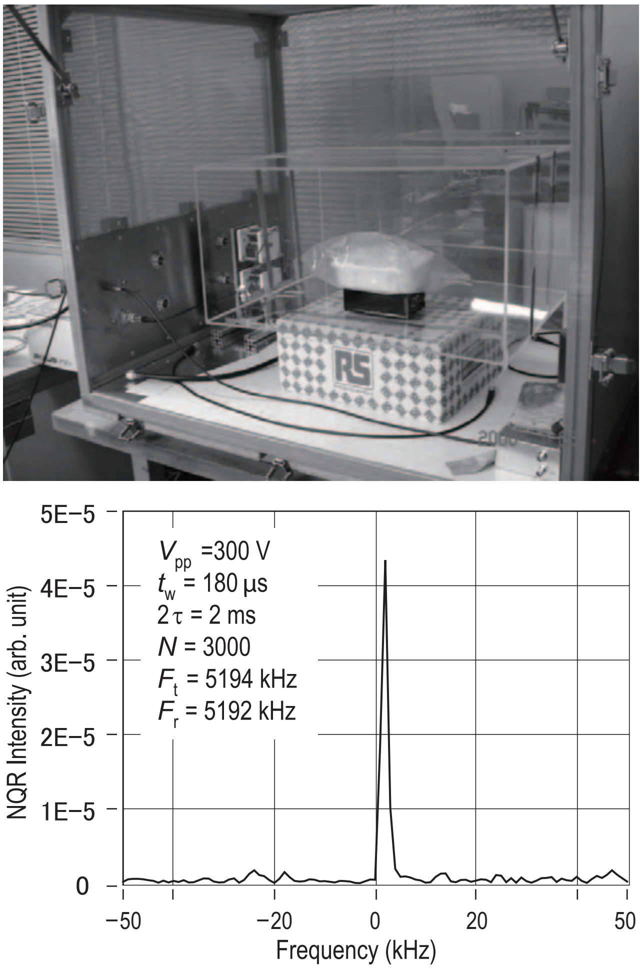

Fig. 6 The one of ν+ lines (5192 kHz at RT) obtained by the FFT method from 300 g of RDX in a large volumetric solenoid. The abscissa is the offset from the reference frequency.

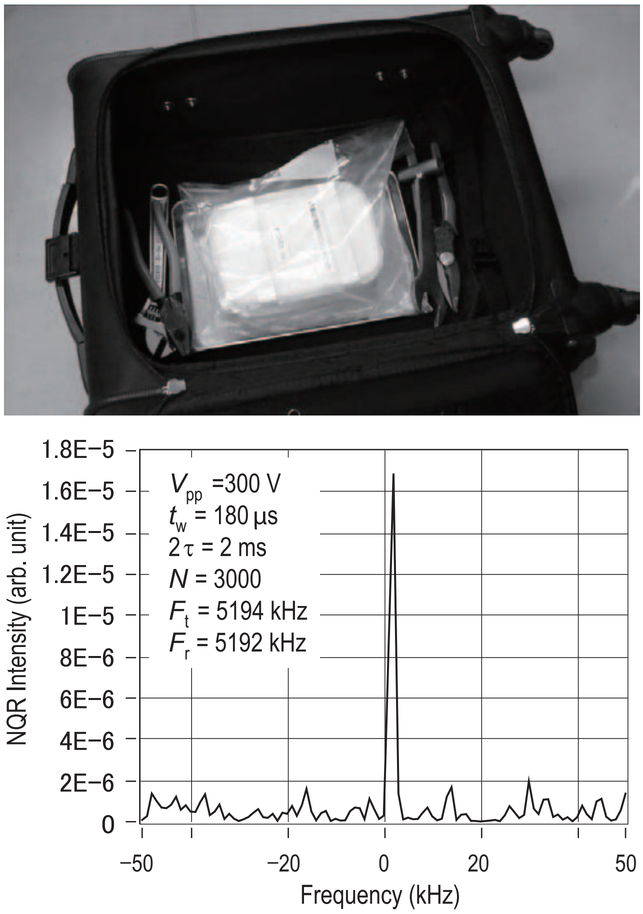

Fig. 7 The one of ν+ lines (5192 kHz at RT) from 300 g of RDX in a baggage which contains a stainless steel tray under the RDX and metallic tools. The abscissa is the offset from the reference frequency.

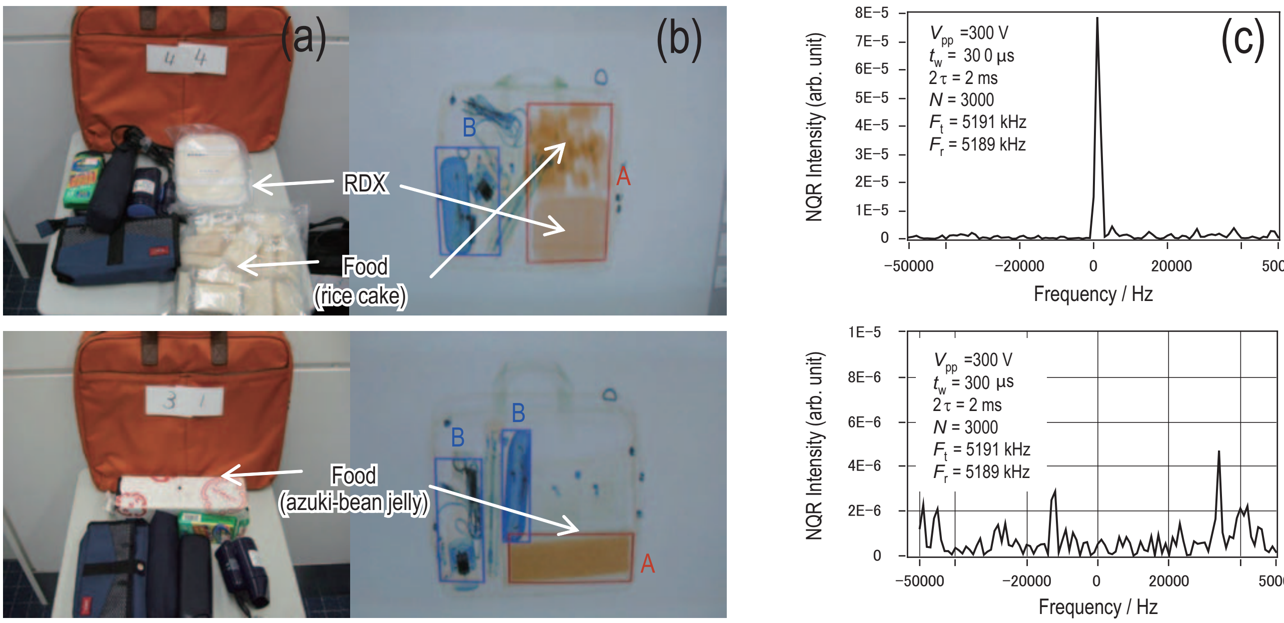

Fig. 8 **(a)** Optical images of the contents of the baggage sample. ***Upper*** : 300 g of deactivated RDX, food (rice cake) with the X−ray transmittance similar to explosives, and travel goods (hair dryer, folding umbrella, glasses in the case, disposable camera and toilet kit). ***Lower*** : Food (azuki−bean jelly) with the X−ray transmittance similar to explosives, and travel goods. **(b)** X−ray images of the baggage sample. The frames of A (a X−ray transmittance in the range of explosives) and B (a X−ray transmittance of metal) were automatically displayed according to the set values. ***Upper and lower*** : The presence of explosive, ***i.e.*** the frame of A, is displayed. **(c)** The results of NQR baggage screening. ***Upper*** : The peak is one of ν+ lines (5192 kHz at RT) from 300 g of RDX in a baggage. ***Lower*** : No NQR peak of RDX was observed and it means nonexistence of RDX.

NQR baggage screening was conducted with 50 kinds of baggage samples for the comparison with explosive detection by X−ray inspection. Fifty kinds of samples were prepared with three types of bags which randomly contain any of the following items ; clothes and travel goods, 300 g or 500 g of deactivated RDX, foods and substances whose X−ray transmittance values were close to those of plastic explosives. The pictures of baggage sample and the automatic judgment of both X−ray image and NQR measurements are shown in Fig. 8 for two baggage samples with and without RDX.

NQR screening correctly judged existence or nonexistence of RDX, on the other hand, X−ray screening responded not only to RDX but also to foods as shown in Fig. 8. By the X−ray inspection, most of the substances whose X−ray transmittance is similar to explosives cannot be distinguished from explosives, resulting in the high value of false positive rate. A false positive rate with NQR screening system was *ca.* one−tenth of that with X−ray inspection system, while the value of false negative rate was as low as that of X−ray inspection. It clearly indicates that the security−check by NQR explosive detection technique is useful for the transportation security.

The most interfering object was a notebook computer in the test. The noise was thought to be caused due to coupling of the receiver coil of NQR system with the circuit in the note PC. The total time required for the measurement was 3 minutes*. Data acquisition to onboard memory was, however, finished within 6 seconds for $N = 3000$. The data processing time can be shortened with a hardware technique like Field Programmable Gate Array (FPGA) [17].

* By the improvement of an algorithm in the software program, the measurement time could be shortened to *ca.* 12 seconds.

4. Summary

The applicability of NQR technique to explosives detection was examined for the transportation security. Prototype shoe and baggage screening system using NQR technique were developed and tested using HMT, $NaNO_2$, or RDX as a target substance. The gradiometer coil for shoe screening detected 300 g of RDX located 3.5 cm above the coil−surface with S/N = *ca*. 15 without an electromagnetic shield. It indicates the feasibility of NQR shoe screening without taking them off. The blind test of NQR baggage screening with large solenoid was conducted and compared with the results of X−ray screening at the security−check gate in the airport. The results confirmed that security check with NQR technique contributed to decrease the false positive rate to *ca.* one−tenth of that of the conventional X−ray inspection. The test also found out, however, that impedance mismatching or coupling noise was caused by existence of metallic objects or electric devices such as a notebook computer. It can be thought for these problems to be solved by the improvement of a protocol in the screening. The typical detection time was 3 minutes and it can be shortened using hardware technique such as FPGA, since most of the time for measurement was software program process and the data acquisition was confirmed to finish within 6 seconds of RF pulse irradiation.

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運輸保安強化のための核四重極共鳴（NQR）を用いた爆薬検知技術の開発

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空港保安検査等に用いられる爆薬検知方法としてはX線を用いて物質の形状及びX線透過率から爆薬の有無を判断する方法が主流であるが，プラスチック爆薬は形状を変化させることが可能であり，また爆薬と同程度のX線透過率をもつ物質も日用品として多く存在するため，誤報率が高いことが課題となっている。一方，核四重極共鳴（NQR）は高周波を照射することで物質に固有の共鳴周波数を測定する方法で物質の特定が可能であり，より正確な爆薬検出技術としての可能性が期待されている。そこで我々は国土交通省の委託の下，空港等の交通機関におけるテロ対策強化を目的として，靴検査を想定した平面検出部および手荷物検査を想定した大容積の検出部を持つNQR爆薬検知装置を開発し，亜硝酸ナトリウム500 g及びRDXの原料であるHMT 414 g，並びに不爆化RDX 300 gを用いた遠隔検出実験によりその有用性を検証した。平面コイルを用いた実験においては，電磁シールドのない環境においてもRDX 300 g程度が検出可能であることを確認した。また，大容積コイルを持つ装置に対しては空港保安検査場において実証試験が実施され，X線検査装置との比較が行われた。RDX等を含む手荷物試験体を用いたブラインド試験の結果からNQR検査における誤報率（false positive rate）はX線検査の10分の１程度であることが実証され，NQRを用いた爆薬検査が交通機関における保安対策強化に有効であることが示された。

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